

(19)



Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11)

**EP 1 123 734 A2**

(12)

**EUROPEAN PATENT APPLICATION**

(43) Date of publication:  
16.08.2001 Bulletin 2001/33

(51) Int Cl.7: **B01J 19/00, B01J 19/24**

(21) Application number: **01102270.4**

(22) Date of filing: **31.01.2001**

(84) Designated Contracting States:  
**AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU  
MC NL PT SE TR**  
Designated Extension States:  
**AL LT LV MK RO SI**

(30) Priority: **03.02.2000 US 496999**

(71) Applicant: **Cellular Process Chemistry Inc.  
Richland, Washington 99352 (US)**

(72) Inventors:  
• **Schwalbe, Thomas, Dr.  
61118 Bad Vilbel (DE)**  
• **Golbis, Klaus, Dr.  
63477 Maintal-Doernigheim (DE)**

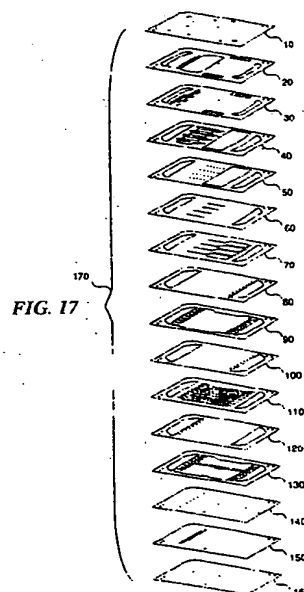
- **Hohmann, Michael, Dr.  
64287 Darmstadt (DE)**
- **Georg, Petra  
60326 Frankfurt (DE)**
- **Oberbeck, Andreas  
56205 Wiesbaden (DE)**
- **Dittmann, Bernd  
65843 Sulzbach (DE)**
- **Stastna, Jiri  
65760 Eschborn (DE)**
- **Oberbeck, Sebastian  
35753 Greifenstein (DE)**

(74) Representative: **Kompter, Hans-Michael  
Spessartring 63  
64257 Darmstadt (DE)**

**(54) Miniaturized reaction apparatus**

(57) A stacked plate chemical reactor in which simple plates, each incorporating no surface features other than an opening, are stacked together. When openings in adjacent plates are properly aligned, a fluid pathway is defined between inlet ports for each chemical reactant and an outlet port for a chemical product. In one embodiment of the invention, sixteen simple plates are stacked to provide a reactor incorporating three heat transfer fluid pathways, two reactant fluid pathways, one product fluid pathway, multiple mixing chambers, multiple reaction chambers, two reactant pretreatment heat exchangers, two reaction chamber heat exchangers, and multiple temperature sensor pathways. Precise dimensional control of the reactant fluid pathway height enables stacked laminar flow paths for the reactants to be achieved, allowing efficient and rapid diffusion mixing to occur. Because the simple plates incorporate no features other than openings, fabrication of such plates is easily achieved. Different reactor designs, having additional reactant pathways, more or fewer heat transfer fluid pathways, more or fewer heat exchangers, more or fewer mixing chambers, more or fewer reaction chambers, and more or fewer sensor pathways can readily be achieved by adding or removing plates from the stack, and/or by changing the pattern and number of openings in the simple plates that are used. The simple plates can be held in the stack during use of the chemical reactor using pressure exerted on opposite

outer simple plates of the stack, or can be permanently joined. A preferred material for the fabrication of the plates is stainless steel, although other materials such as glass, plastic, and other metals can alternatively be used, which are compatible with the selected reactants and the desired product.

**EP 1 123 734 A2**

**Description****Field of the Invention**

5 [0001] This invention generally relates to a miniaturized chemical processing apparatus, and more specifically, to a miniaturized chemical processing apparatus assembled from stacked plates that cooperate to provide fluid channels for conveying reactants and other fluids.

**Background of the Invention**

10 [0002] Methods of controlling and optimizing processes for producing chemical compounds are well known. The control of parameters such as temperature, pressure, mixing conditions, relative volumes of the reactants, and the use of catalysts are generally well understood. Traditionally, newly discovered chemical compounds and processes involving either the production of such compounds, or processes involving the use of such compounds, have been initially  
15 carried out by researchers in "bench-scale" environments. Particularly promising chemicals or processes may ultimately be produced in quantity by application to industrial scale processes. Often, problems are encountered in scaling up a process from the laboratory to industrial scale production.

[0003] Problems associated with moving from bench-scale production to industrial scale production often involve changes in process conditions between the bench-scale environment and the industrial environment. For example,  
20 the temperature of the reactants within a small beaker or flask in a laboratory is much easier to keep constant than the temperature within a production tank having a capacity of hundreds of liters, as is often the case in a chemical processing plant. Variations in other process conditions within a large tank are also more difficult to control, and frequently effect the quality and yield of the desired product.

[0004] Another aspect of laboratory development of processes to produce chemical compounds is that often poten-  
25 tially dangerous chemicals are used to create the desired product. Fires and explosions in research laboratories and concomitant injury to personnel and property are well known risks in the chemical research industry. The risks are not limited only to research only, as industrial chemical production facilities also may experience fires and explosions related to chemical production using dangerous chemicals. Often, due to the quantities of chemicals used in industrial scale processes, such accidents are significantly more devastating in an industrial setting than similar accidents in a  
30 research setting.

[0005] Recently, much attention has been directed to the use of micro-scale reactors for both development and production of chemical processes. These types of reactors offer several advantages. As noted above, the control of chemical processes within very small reactors is much simpler than control in a large-scale production tank. Once a reaction process has been developed and optimized in a micro-scale reactor, it can be scaled up to industrial production  
35 level by replicating the micro-scale reactors in sufficient quantity to achieve the required production output of the process. If such reactors can be fabricated in quantity, and for a modest cost, industrial quantities of a desired product can be manufactured with a capital expenditure equal to or even less than that of a traditional chemical production facility. An additional benefit is that because the volume of material in each individual reactor is small, the effect of an explosion or fire is minimized, and with proper design, an accident in one reactor can be prevented from propagating to other  
40 reactors.

[0006] Safety in the research setting is also improved, as such reactors generally require less exposure to hazardous substances and conditions by research personnel than traditional "wet-chemistry," which typically requires that the researcher physically handle chemicals in a variety of glass containers, often in the presence of an open flame and/  
45 or other heat sources. Any accident in such an environment is likely to increase the risk that the researcher will be exposed to hazardous chemicals, as well as the risk of causing significant damage to the laboratory. In contrast, small scale or microreactors can be designed as self-contained units that minimize the researcher's potential exposure to chemical substances. Since when using a microreactor, the researcher is not required to physically manipulate containers of chemical materials to carry out a desired reaction, the reactor can be located in an area so that if an accident should occur, any resulting fire or explosion can be relatively easily contained.

[0007] Another area in which microreactors offer an advantage over conventional chemical process development and production is in the mixing of reactants. A mixing channel of the proper scale encourages a laminar flow of the reactants within the channel and is readily achievable in a microreactor. A laminar flow enhances mixing by diffusion, which eliminates the need to expend energy to physically stir or agitate the reactants and is an extremely fast and efficient mixing technique.

50 [0008] Microreactors particularly offer great promise to the pharmaceutical industry, which engages in chemical research on many new chemical compounds every year, hoping to find a drug or chemical compound with desirable and commercially valuable properties. Enhancing the safety and efficiency of such research is valuable in and of itself. And, when coupled with the potential that these reactors offer for eliminating the problems of moving from bench-scale

production to industrial production, it will be apparent that a microreactor suitable for use in carrying out a variety of chemical processes and having an efficient and low-cost design will be in high demand.

[0009] Several different designs for microreactors have been investigated. For example, such reactors are described in U.S. Patent No. 5,534,328 and U.S. Patent No. 5,690,763 (both listing Ashmead as the inventor). These patents describe reactor structures for chemical manufacturing and production, fabricated from a plurality of interconnected layers. Generally, each layer has at least one channel or groove formed in it and most include orifices that serve to fluidly connect one layer to another. These layers are preferably made from silicon wafers, because silicon is relatively inert to the chemicals that may be processed in the reactor, and because the techniques required to mass produce silicon wafers that have had the required channels and other features etched into their surfaces are well known.

[0010] A disadvantage of the reactors described by Ashmead stems from the rather expensive and complicated process for manufacturing the devices. While silicon wafer technology is advanced to the state that wafers having desired surface features can readily be mass produced, the equipment required is capital intensive, and unless unit production is extremely high, the substantial costs are difficult to offset. While Ashmead does suggest that other materials can be used to fabricate the layers, such as metal, glass, or plastic, the surface features required (grooves, channels, etc.) must still be formed in the selected material. The particular surface features taught by Ashmead require significant manufacturing steps to fabricate. For instance, while forming an opening into a material is relatively easy, forming a groove or channel that penetrates only part way through the material comprising a layer is more difficult, as the manufacturing process must not only control the size of the surface feature, but the depth, as well. When forming an opening that completely penetrates through a material comprising a layer, depth control does not need to be so precisely controlled. Ashmead teaches that not only openings that completely penetrate the layers are required, but also that surface features (grooves/channels) that do not completely penetrate the individual layers are required. Hence, multiple processing steps are required in the fabrication of each layer, regardless of the material selected. Accordingly, it would be desirable to develop a microreactor comprising layers that do not require such detailed fabrication.

[0011] A patent issued to Bard (U.S. Patent No. 5,580,523) describes a modular microreactor that includes a series of fluidly connected modules, each module having a particular function (fluid flow handling and control, mixing, chemical processing, chemical separation, etc.). Bard specifically teaches that the plurality of modules are mounted laterally on a support structure, and not stacked, as disclosed by Ashmead. In a preferred embodiment of Bard, silicon wafer technology is again used to etch channels and/or other features into the surface of a silicon wafer. Other disclosed fabrication techniques include injection molding, casting, and micromachining of metals and semiconductor substrates. Again, the processing required to fabricate the individual modules goes beyond merely forming a plurality of openings into each component. Furthermore, the lateral layout of the reactor described by Bard requires a larger footprint than a stacked plate reactor. In Bard's reactor, the more modules added, the larger the footprint of the entire reactor. In contrast, when additional plates are added to a stacked plate reactor, the footprint of the reactor does not change, which can be a distinct advantage, as in many work environments the area an apparatus occupies on a work bench or floor is more valuable than the vertical height of the apparatus. It would be desirable to provide a reactor design that has a minimal footprint, while still providing the flexibility to add components to customize the reactor for a particular process.

[0012] In U.S. Patent No. 5,961,932 (Ghosh), a reactor is described that is formed from a plurality of ceramic layers, which are fluidly connected, at least one layer including a permeable partition. In particular, Ghosh describes the desirability of sizing fluid channels appropriately to provide for laminar flow and mixing via diffusion, rather than mixing via turbulence. In his preferred embodiment, Ghosh describes that channels, chambers, and passageways are formed in each layer. The particular process Ghosh describes to accomplish this task involves fabricating the layers from "green" or uncured ceramic, which once shaped as desired, must be sintered. Significantly, the sintering process changes the size of the ceramic layer, so that the sizes of the features formed into the ceramic layer in the initial stages of production are not the sizes of the features in the finished product. It would be desirable to provide a reactor design in which the dimensions of the individual components can be rigidly controlled during fabrication; and not subject to shrinkage, which can negatively effect the dimensions of the finished reactor. This object is particularly important when a reactor design focuses on achieving a laminar flow, because precise dimensional control of fluid pathways in the reactor must be maintained to achieve a consistent laminar flow.

[0013] In all of these prior art reactors, relatively complicated manufacturing techniques are required. The manufacture of layers of silicon material requires a large capital investment. Sintering of a ceramic material requires the precise control of the shrinkage process, or individual components of a desired size cannot be achieved. In all cases, the prior art teaches that complicated structures (for example, fluid channels and reaction chambers) must be etched or otherwise fabricated in each layer. Additionally, orifices or passages also need to be formed in each layer, so that fluids can move between adjacent layers of the reactor. Thus, a series of different manufacturing steps typically must be performed for each layer. It would be desirable to provide a reactor design offering the advantages described above, that is relatively simple to manufacture, so as to minimize capital investment in scaling up production from the laboratory to the industrial level. It is therefore an aim of this invention to provide a micro-scale reaction apparatus that can be

economically manufactured, can maintain a desired relatively narrow temperature range for a process, has a relatively modest footprint, and can provide efficient diffusion mixing using a precisely controlled laminar flow.

### Summary of the Invention

**[0014]** In accord with the present invention, a reactor is defined for reacting one chemical with at least one other chemical, for the purpose of forming a chemical product. The reactor includes a plurality of simple plates, each simple plate having at least one opening formed therein, the simple plates being stacked together to form a plurality of layers and arranged so that at least one opening in each simple plate overlaps at least one other opening in an adjacent simple plate, thereby forming at least one pathway between at least some of the layers.

**[0015]** Preferably, openings within different layers align so as to form at least one inlet port and at least one outlet port, for the receipt and discharge of chemicals, and to form at least one pathway for conveying chemicals to be processed. At least one pathway is formed that is in fluid connection with the inlet and outlet ports, and each simple plate has at least one opening formed in it.

**[0016]** A material from which the simple plates are fabricated is selected for compatibility with the chemical process. In one embodiment, the simple plates are formed from a material selected from the group consisting of crystalline wafers, ceramics, glasses, polymers, composite materials, and metals. Preferably, if formed from a metal, stainless steel is used. The material of the crystalline wafer is selected from the group consisting of silicon and germanium.

**[0017]** It is also preferable that the reactor accommodate a plurality of operations, including temperature control, control of chemical residence time, chemical mixing, and chemical reacting. Temperature control is achieved using a combination of one or more temperature sensors and one or more heat exchangers. Preferably, chemical mixing is carried out by employing pathways sized so that a reactant achieves a stacked laminar flow with respect to at least one other reactant.

**[0018]** In a reactor adapted for processing at least two reactants to form a desired chemical product, an inlet opening for each of the reactants and an outlet opening for the chemical product is provided in at least one of two outer simple plates. An intermediate simple plate is included for mixing the reactants and has at least one opening in fluid communication with each inlet opening and the outlet opening.

**[0019]** Generally, at least one heat transfer fluid inlet port is included in at least one of the outer simple plates, so that at least one heat transfer fluid can be introduced into the chemical reactor. Each heat exchanger is defined by an opening in a different intermediate simple plate. The opening is in fluid communication with the heat transfer fluid inlet and outlet ports and is disposed between adjacent simple plates.

**[0020]** Preferably, each heat exchanger is used to modify the temperature of at least one of the reactants and/or the chemical product. The heat exchangers can be used to modify a temperature of one of two reactants such that they are at different temperatures.

**[0021]** The chemical reactor typically includes a plurality of intermediate simple plates, and the openings in these plates define a first fluid path for a first of the at least two reactants, and a second fluid path for a second of the at least two reactants. Preferably, the plurality of intermediate simple plates define an inter-digital-mixer that separates and aligns the first fluid path and the second fluid path into a plurality of individual fluid paths. The plurality of individual fluid paths are then joined in a laminar flow pathway to provide a stacked laminar flow of the first and second reactants. The stacked laminar flow enables mixing of the reactants to be achieved by diffusion mixing. Preferably, a height of the joined fluid paths is reduced to enhance the diffusion mixing. In at least one embodiment, the height of each individual stacked laminar flow is reduced to less than 50 micrometers.

**[0022]** In one embodiment, a width of the laminar flow pathway is increased, so that a flow rate of a fluid in the pathway remains constant as the height of the pathway is reduced. Preferably, when the inter-digital-mixer separates and aligns the first fluid path and the second fluid path into a plurality of individual fluid paths, a pressure drop for each of the individual fluid paths are substantially equivalent. The inter-digital-mixer also ensures that when the first fluid path and the second fluid path are separated and aligned into a plurality of individual fluid paths, so that each individual fluid path enters the at least one opening in which the individual fluid paths are joined, from the same side.

**[0023]** In one embodiment, the openings in the plurality of intermediate simple plates share common shapes and sizes to the extent possible, to minimize fabrication costs. Preferably, all the simple plates are chamfered at one corner to provide a reference when assembling the simple plates to form the chemical reactor.

**[0024]** Because temperature control of reactants and the resulting product is critical to yield and purity, the chemical reactor preferably includes at least one temperature sensor to monitor a temperature of the product or at least one of the reactants. A temperature sensor is disposed in at least one of the outer simple plates, and another temperature sensor is disposed in at least one of the plurality of intermediate simple plates.

**[0025]** The thickness of the outer simple plates is about 3 millimeters, and that of the plurality of intermediate simple plates is at least about 0.2 millimeters, but not more than about 0.6 millimeters.

**[0026]** In one embodiment, the simple plates are removably held together in the stack by an applied compressive

force. In such an embodiment, a housing provides the compressive force, producing a pressure acting on the outer simple plates. The mean surface roughness of the plates should be less than about 1 micrometers, and the simple plates should be substantially free of scratches. The pressure should be greater than or equal to 50 Newtons per square centimeter. In another embodiment, the simple plates are permanently joined. When permanently joined, the mean surface roughness of the plates is preferably less than about 5 micrometers. Permanent joining can be achieved using diffusion welding or vacuum soldering.

[0027] Preferably, when the thickness of the intermediate simple plates that are adjacent to a heat exchanger is about 0.3 millimeters. When a series of openings in the simple plates of the chemical reactor defines a fluid path for a heat transfer fluid that flow through more than one heat exchanger, the flow rate and fluid pressure of the heat transfer fluid within each such heat exchanger are substantially.

[0028] Another aspect of the present invention is directed to a method for producing stacked plate reactor, which includes steps generally consistent with the apparatus described above.

#### Brief Description of the Drawing Figures

[0029] The foregoing aspects and many of the attendant advantages of this invention will become more readily appreciated as the same becomes better understood by reference to the following detailed description, when taken in conjunction with the accompanying drawings, wherein:

FIGURE 1 is a plan view of a top simple plate of a preferred embodiment for a chemical reactor in accord with the present invention, including openings for reactants, heat transfer media inlets and outlets, and an opening for a temperature sensor;

FIGURE 2 is a plan view of the second simple plate of the preferred reactor, showing a plurality of heat exchanger manifolds, a first reactant opening, a second reactant distributor, and a first heat exchanger;

FIGURE 3 is a plan view of the third simple plate of the preferred reactor, illustrating a plurality of heat exchanger manifolds, a first reactant distributor, and a second reactant opening;

FIGURE 4 is a plan view of the fourth simple plate of the preferred reactor, showing two heat exchanger manifolds, an inter-digital-mixer for the reactants, and a second heat exchanger;

FIGURE 5 is a plan view of the fifth simple plate of the preferred reactor, showing two heat exchanger manifolds, a plurality of openings for the reactants, and a second heat exchanger;

FIGURE 6 is a plan view of the sixth simple plate of the preferred reactor, showing two heat exchanger manifolds, and a plurality of reactant fluid channels;

FIGURE 7 is a plan view of the seventh simple plate of the preferred reactor, showing two heat exchanger manifolds, a plurality of reactant fluid channels, and a plurality of mixing chambers for the reactants;

FIGURE 8 is a plan view of the eighth simple plate of the preferred reactor, showing two heat exchanger manifolds and a plurality of product openings;

FIGURE 9 is a plan view of the ninth simple plate of the preferred reactor, showing two heat exchanger manifolds, a third heat exchanger, and a plurality of product openings;

FIGURE 10 is a plan view of the tenth simple plate of the preferred reactor, showing two heat exchanger manifolds and a plurality of product openings;

FIGURE 11 is a plan view of the eleventh simple plate of the preferred reactor, showing two heat exchanger manifolds, and a plurality of reaction channels;

FIGURE 12 is a plan view of the twelfth simple plate of the preferred reactor, illustrating two heat exchanger manifolds, and a plurality of product openings;

FIGURE 13 is a plan view of the thirteenth simple plate of the preferred reactor, illustrating two heat exchanger manifolds, a plurality of product openings, a plurality of temperature sensor openings, and a fourth heat exchangers that is separated into an upper section and a lower section;

FIGURE 14 is a plan view of the fourteenth simple plate of the preferred reactor, illustrating a plurality of product openings and a plurality of temperature sensor openings;

FIGURE 15 is a plan view of the fifteenth simple plate of the preferred reactor, illustrating a product channel and a plurality of temperature sensing openings;

FIGURE 16 is a plan view of the sixteenth and bottom simple plate of the preferred reactor, illustrating a product withdrawal opening and a plurality of temperature sensor openings;

FIGURE 17 is an exploded isometric view of the preferred reactor, illustrating how all sixteen simple plates are stacked;

FIGURE 18A is an exploded isometric view of the first six simple plates of the preferred reactor, illustrating a fluid path for a first reactant;

FIGURE 18B is an exploded isometric view of the first six simple plates of the preferred reactor, illustrating a fluid

path of a second reactant;

FIGURE 18C is an exploded isometric view of simple plates seven through sixteen of the preferred reactor, illustrating the combined fluid paths of the first and second reactants after they have been mixed, and then through the balance of the preferred reactor;

FIGURE 19A is an exploded isometric view of the first two simple plates of the preferred reactor, illustrating a fluid path for heat transfer media servicing the first heat exchanger;

FIGURE 19B is an exploded isometric view of the first four simple plates of the preferred reactor, illustrating a fluid path for heat transfer media servicing the second heat exchanger;

FIGURE 19C is an exploded isometric view of the first thirteen simple plates of the preferred reactor, illustrating the fluid paths for heat transfer media servicing heat exchangers three and four;

FIGURE 20 is a cross-sectional view of simple plates five through seven of the preferred reactor, illustrating both the relative thickness of these plates and a fluid path for both reactants through the inter-digital-mixer mixer and the mixing areas;

FIGURE 21 is a schematic view of selected portions of simple plates five through seven of the preferred reactor, illustrating how the inter-digital-mixer provides a stacked laminar flow of the reactants in the mixing areas of the seventh simple plate;

FIGURE 22 is a schematic view of selected portions of an inter-digital-mixer that does not enable the desired stacked laminar flow of reactants; and

FIGURE 23 is a schematic view of selected portions of a preferred design for an inter-digital-mixer that does enable the desired stacked laminar flow of reactants.

#### Description of the Preferred Embodiment

[0030] The present invention is a miniaturized chemical reaction apparatus fabricated from a plurality of simple plates. Unlike prior art stacked layer chemical reactors that require relatively complicated surface features, such as grooves or channels that do not penetrate the component to be formed into each layer, the simple plates employed in the present invention require no more than an opening be formed through each plate. Machining or stamping openings into a flat plate is significantly less complicated than the silicon etching, injection molding, and ceramic molding/sintering processes described in the prior art for producing the surface features that the prior art uses to channel fluid flow. Yet the relatively simple technique of forming openings in a flat plate can be used to achieve a very useful chemical reactor, if the openings are properly placed, and the plates are properly configured and stacked so that the openings in the plates cooperate to convey fluids through the apparatus.

[0031] In the following description and the claims that follow, it will be understood that the term "simple plate" means a plate that has substantially planar opposed surfaces, e.g., a flat sheet of material. The simple plates used in the embodiments of the present invention disclosed herein are all generally rectangular and are characterized by having one or more openings that pass completely through the simple plate. Thus, the term "simple plate" as used herein and in the claims should be understood to mean a plate that does not include any etchings, grooves, or channels that do not completely penetrate the plate.

[0032] The term "groove," as used herein, should be understood to mean a surface feature that has been formed into the surface of an object, that does not penetrate completely through the object, and applies to components of prior art chemical reactors. The term "crystalline wafer," as used herein and in the claims that follow, means a material that has a crystalline structure and has been sliced into wafer-like components. Silicon and germanium are examples of materials employed for producing such crystalline wafers; however, it is not necessary for the material to be a semiconductor to comprise a crystalline wafer. Also, it is not necessary for the material to be a single element such as silicon or germanium, but rather such a material can be a mixture of several elements that together form a material, which can be fabricated into crystalline wafers. The fabrication techniques commonly used in the semiconductor industry to form substrate wafers can be employed to produce crystalline wafers.

[0033] In a preferred embodiment, the simple plates are formed of a high quality stainless steel, and standard metal working techniques such as stamping and/or milling are used to fabricate the simple plates. It should be understood that a variety of other materials can be used to fabricate simple plates. Metals other than stainless steel can alternatively be used, as well as other materials, such as glass, plastic, or a combinations of these materials. Crystalline wafers are another alternative material from which to form the simple plates. The use of other materials will be accompanied by fabrication techniques appropriate to the specific type of material, such as injection molding for plastic materials. The material used to fabricate the plates must be considered in light of the chemical properties of the reactants used in a particular reaction. Stainless steel is a relatively chemically inert material, and is an appropriate material for many chemical reactants. Tantal alloys and silver alloys are also expected to be useful. Hydrofluoric acid is a chemical that is extremely corrosive to metals and glass. Special plastic materials are appropriate when the desired reaction involves hydrofluoric acid. Those of ordinary skill in the art of chemical processing will readily understand how the choice of

reactants necessitates an appropriate material be selected for fabricating the simple plates of the reactor.

[0034] A preferred embodiment of the present invention, as described below, represents a design that has been optimized for a liquid/liquid phase reaction involving two reactants. It should be understood that the underlying concept of the present invention, i.e., a reactor formed of a stack of plates incorporating only openings, can be applied to many other types of reactions, such as liquid/gas, gas/gas, liquid/solid, or gas/solid. As will be described in detail below, the preferred embodiment includes four heat exchangers; three heat transfer media pathways, and two reactant fluid pathways. However, it should similarly be understood that similar stacked plate reactors can be easily designed to include more or fewer heat exchangers, more or fewer heat transfer media pathways, and more reactant pathways.

[0035] The disclosed preferred reactor has been optimized for processing two component liquid/liquid reactions that generally require only temperature controls. However, it should be understood that other types of reactions, requiring additional processing controls, can be processed in a stacked simple plate reactor in accord with the present invention, if the reactor is optimized for that control parameter. For example, reactors can readily be designed to incorporate magnetic, piezoresistive, piezoelectric, shape memory, radioactive, catalytic, and electrostatic control parameters.

[0036] The plurality of stacked simple plates enables a reactor to be constructed that performs from one to all of the following functions: reactant conditioning, control of reactant supply, thermal pre-treatment, combination and mixing of reactants under controlled thermal conditions, intermediate thermal treatment, post-procedural isothermal containment, post-procedural thermal treatment of reactant products, and product separation. In particular, simple plates can readily be designed and fabricated in which the dimensional characteristics of the reactant fluid passages formed by the interconnected openings of the simple plates provide for a stacked laminar flow of the reactants. Such a stacked laminar flow ensures that a particularly efficient type of mixing, referred to as diffusion mixing, can occur.

[0037] The quality of the interconnections between the simple plates is of great importance, since the interconnections must be free of gaseous and liquid leakage. This requirement is achieved through a combination of specially prepared surfaces and use of simple plates that are fabricated to close tolerances. The individual simple plates can be assembled by pressure fitting (using clamps or a housing that encloses the simple plates and applies a compressive force to the outer plates), or individual simple plates can be permanently assembled using diffusion welding technology, vacuum soldering, or other suitable techniques for joining the simple plates together.

[0038] The pressure fitting technique has the advantage of allowing a reactor to be built using specific simple plates that can readily be disassembled so that the reactor design can be changed by adding or removing simple plates. In this manner, the same simple plates can be used in more than one reactor to effect different chemical processes. However, if the simple plates are assembled using pressure fitting, very good control of the surface finishes is required, with almost no scratches on the surface of the simple plates, and a mean surface roughness less than 1  $\mu\text{m}$ . The pressure that should be applied to maintain a stack of simple plates that have been fabricated from metals into a reactor, to prevent gas or liquid leakage, is preferably about 50 Newtons/cm<sup>2</sup>.

[0039] Successful diffusion welding to join metallic simple plates also requires a substantially scratch free surface, although the mean surface roughness can be increased up to 5  $\mu\text{m}$ . In diffusion welding, the simple plates are pressed together and heated to about 1000° C in a vacuum or inert atmosphere. At such temperatures, ions from each surface diffuse across the surface boundary layers, thus joining the surfaces.

[0040] Vacuum soldering is a technique that requires a mean surface roughness of less than 5  $\mu\text{m}$ , although more scratches can be tolerated than in diffusion welding. The simple plates are first coated with a thin film (3-5  $\mu\text{m}$ ) of silver, either by sputtering, vapor deposition, or electrical deposition. Other metallic films, such as gold or copper, can also be used. The simple plates are then heated in a vacuum to about 900° C. The silver liquefies, filling any voids due to scratches or surface irregularities, and bonds the simple plates together to form a reactor.

[0041] It should be noted that when the reactor is assembled using diffusion welding or vacuum soldering, a superior bond can be obtained by minimizing the surface area that is to be bonded. Thus, simple plates that incorporate one or more openings occupying a significant portion of the surface area of the simple plates can be more efficiently bonded with either of these two techniques than simple plates with few or small openings that comprise only a small portion of their total area.

[0042] Preferably, any stacked simple plate reactor should have the ability to maintain a desired narrow temperature range within the reactor, so that reaction dynamics can be closely controlled. In a preferred embodiment, the reactant and heat transfer media enter the stacked simple plate reactor via vertically oriented fluidic channels. Reacted product and spent heat transfer media exit the reactor via similarly disposed vertically oriented fluidic channels. The chemical processing operations occur in horizontally disposed channels within the reactor. It should be noted that the use of the term channel when used in conjunction with a stacked simple plate reactor should not be construed to mean that such a channel corresponds to a groove formed into the surface of an individual plate. While each individual simple plate only has openings and no grooves, channels or other fluid pathways are easily obtained in a stacked simple plate reactor. To form a channel, an elongate narrow opening is formed in one simple plate and that simple plate is sandwiched between two simple plates that do not have a corresponding elongate opening. The top of the channel is defined by the upper simple plate, the sides of the channel are defined by the sides of the opening formed in the middle simple

plate, and the bottom of the channel is defined by the bottom plate. Thus, the depth of the channel is the same as the thickness of the middle simple plate. Fluid pathways between adjacent simple plates within a stacked simple plate reactor are created when openings through the stacked simple plates are aligned.

**[0043]** To achieve precise control of the desired reactions, the stacked simple plate reactor preferably includes a control circuit with several temperature sensors, as well as flow controls for a heat transfer media that is circulated through the reactor. The sensors may be disposed outside of the stacked simple plate reactor, but are preferably disposed either within the stacked simple plate reactor, or in the reactor housing, if such housing is provided. Note that if the simple plates are permanently joined, or clamps are used to press the simple plates together, then a reactor housing is not required. The control circuit may similarly be mounted externally, or disposed within the stacked simple plate reactor, or within the reactor housing (if such a housing is provided).

**[0044]** The fluidic system of the stacked simple plate reactor is preferably characterized by having a small pressure drop across the entire system. Furthermore, potential clogging problems are minimized by having few constrictions within the reactor, by introducing as few sharp directional flow changes as possible, by maintaining a small inner volume (about 1 ml), and by enabling rapid diffusion mixing in the mixing portion of the reactor. Preferably, fluidic channel geometries range from 100-500  $\mu\text{m}$ , especially with respect to reactant fluid pathways (the dimensions of any heat transfer media pathways are less critical), and the walls separating the heat transfer media from the reactants or product should be of similar scale, to enable rapid heat transfer. As discussed above, several materials can be used to fabricate a stacked simple plate reactor; however, simple plates that are adjacent to openings in those simple plates comprising heat exchangers are preferably fabricated from a material that has good thermal conductivity. However, if the dimensional thickness of each plate adjacent to a heat exchanger is small, on the order of 0.3 mm, the effect of the thermal conductivity of different materials is negligible.

**[0045]** In general, the openings in each simple plate of a stacked simple plate reactor correspond to a fluid pathway established by stacking a plurality of simple plates, such that openings in simple plates above and below overlap, thereby allowing fluids to move throughout the reactor. Openings may also correspond to passageways for sensors, particularly temperature sensors. Preferably, to maximize heat transfer, the fluid flow directions of the heat transfer media within openings defining a heat exchanger are opposite to the direction of reactant flow.

**[0046]** FIGURES 1-16 illustrate simple plates 1-16, with FIGURE 1 representing a top simple plate 10, and FIGURE 16 illustrating a bottom simple plate 160. The preferred reactor includes these 16 simple plates, stacked one on top of another, in 16 layers. In the Figures, the Figure number corresponds to the layer or plate of the reactor for FIGURES 1-16. Thus, FIGURE 1 represents the first layer; FIGURE 2 represents the second layer, and so on. FIGURE 17 shows the orientation of the 16 simple plates stacked one on top of another. FIGURES 18A-18C show the reactant fluid paths (for Reactants A and B) through the stacked plate design. FIGURES 19A-19C illustrate the fluid paths of heat transfer media to each heat exchanger within the stacked plate design. FIGURES 20-23 illustrate further details related to achieving the desired stacked laminar flow, which is critical to the success of this design, as such laminar flow enables rapid and efficient diffusion mixing to occur.

**[0047]** With reference to FIGURE 1, a preferred shape for the simple plates is generally rectangular, though other shapes, such as circular disks, can also be used. It should be noted that all plates preferably include a chamfered edge, to aid in the stacking of the plates in the proper orientation relative to each other. If one plate were oriented in the wrong direction, it is likely that at least one, if not all, of the fluid paths through the reactor would be blocked, disrupted, or improperly joined, and the reactor would likely not be functional. Thus, top plate 10 has a chamfer 11, in the upper left-hand corner. It is not relevant at which corner the chamfer is disposed, as long as all the chamfers on all the succeeding plates are orientated and positioned in the same way, to ensure that once assembled, continuous fluid paths for reactants and heat transfer media are achieved in the stacked simple plate reactor. Similarly, a different type of indexing feature than a chamfered edge could be used as an alignment reference, such as a slot or other mark or shape variation consistently represented on each simple plate. The chamfered corner is preferred, as it is a feature easily incorporated into a simple plate, and chamfers are already recognized as alignment indicators, and widely used in the semiconductor industry on silicon wafers.

**[0048]** In addition to chamfer 11, top simple plate 10 incorporates a plurality of openings for heat transfer media, reactants, and a temperature sensor. Top simple plate 10 incorporates three heat transfer media inlets 12a, 14a, and 16a (for heat transfer media A-C, respectively) and three heat transfer media outlets corresponding to each heat transfer media inlet; including an outlet 12b for a heat transfer media A, an outlet 14b for a heat transfer media B, and an outlet 16b for a heat transfer media C. Two reactant inlet ports are also provided, an opening 15 for a Reactant A, and an opening 17 for a Reactant B. A temperature sensor opening 19 is also provided and will be aligned with corresponding temperature sensor openings in other simple plates, thus forming a temperature sensor passageway within the reactor in which a temperature sensor can be inserted and disposed at a desired location within the stacked simple plate reactor.

**[0049]** FIGURE 2 illustrates the second layer of the preferred reactor, showing a second simple plate 20. As described above, second simple plate 20 also includes chamfer 11. Openings corresponding to every opening in simple plate 10 are also providing in simple plate 20; however, it should be noted however that almost all of the openings are shaped



differently. Heat transfer media A, entering the reactor from inlet 12a in simple plate 10, passes through second simple plate 20 via a heat transfer media A intake manifold 22a. After progressing through subsequent simple plates of the preferred reactor, heat transfer media A again passes through simple plate 20, this time via a heat transfer media A exhaust manifold 22b. From that point, heat transfer media A exits the preferred reactor via outlet 12b in top simple plate 10.

[0050] In second simple plate 20, heat transfer media B services a first heat exchanger 24. Heat transfer media B enters first heat exchanger 24 through inlet 14a in top simple plate 10, and flows from first heat exchanger 24 via outlet 14b in top simple plate 10. Reactant A flows into a Reactant A distributor 25 in second simple plate 20, while Reactant B, passes through second simple plate 20 via Reactant B opening 17. Heat transfer media C flows through second simple plate 20 using a heat transfer media C intake manifold 26a, passing through subsequent simple plates and then returning via a heat transfer media C exhaust manifold 26b. Temperature sensor A (not shown) passes through second simple plate 20 within temperature sensor opening 19.

[0051] The fluid paths of heat transfer media A-C, and the purposes of the four separate heat exchangers in the preferred stacked simple plate reactor will be discussed in detail below, with respect to FIGURES 19A, 19B, and 19C. Briefly, the purpose of first heat exchanger 24 in simple plate 20 is to modify the temperature of Reactants A and B before they are combined. In many reactions, it is desirable for the reactants to be at the same temperature prior to mixing the reactants. However, there are certain reactions for which it is beneficial for a Reactant A and a Reactant B to be brought to different temperatures prior to mixing. It is envisioned that a stacked simple plate reactor can be designed to achieve this goal by changing the openings through the simple plates described above to provide for an additional heat exchanger. Thus, separate heat exchangers can be provided and used to separately modify the temperatures of Reactants A and B.

[0052] FIGURE 3 provides details of the passages in the third layer of the preferred reactor. A third simple plate 30 includes chamfer 11. Heat transfer media A flows through third simple plate 30 via heat transfer media A intake manifold 22a and out through heat transfer media A exhaust manifold 22b. Note that heat transfer media B does not flow through the third layer of the preferred reactor, as heat transfer media B feeds first heat exchanger 24 in second simple plate 20 in the second layer of the preferred reactor and is then exhausted through outlet 14b in top simple plate 10. Heat transfer media C flows through the third layer via heat transfer media C intake manifold 26a, and also via heat transfer media C exhaust manifold 26b. Reactant A flows through third simple plate 30 using a plurality of Reactant A fluid openings 35, which are fed from Reactant A distributor 25 in second simple plate 20. Reactant B enters the third layer of the preferred reactor via Reactant B opening 17 of second simple plate 20, and then flows into a Reactant B distributor 37 in third simple plate 30. Third simple plate 30 also includes temperature sensor opening 19.

[0053] FIGURE 4 provides details of the fourth layer of the preferred reactor, showing a fourth simple plate 40, which has chamfer 11. Heat transfer media A flows through fourth simple plate 40 via a heat transfer media A manifold 42a and out through a heat transfer media A exhaust manifold 42b. It should be noted that the shape of the heat transfer media A intake manifold and exhaust manifold in simple plate 40 have changed in shape from the corresponding heat transfer media A intakes 22a and exhaust manifolds 22b in second simple plate 20 and third simple plate 30. The functional significance of this shape change is to reduce the overall pressure drop in the reactor. The slight curve on the outside edges of intake manifold 42a and exhaust manifold 42b has been included to optimize the fluid flow of heat transfer media A within the preferred reactor. It should also be noted that because intake manifold 42a and exhaust manifold 42b are larger in size than intake manifold 22a and exhaust manifold 22b, the surface area of the simple plate is reduced. As noted above, a smaller surface area results in a superior bond if diffusion welding or vacuum soldering is used to assemble the simple plates in the stack.

[0054] Fourth simple plate 40 of FIGURE 4 incorporates a second heat exchanger 46, which is serviced via heat transfer media C intake manifold 26a and heat transfer media C exhaust manifold 26b of simple plate 30. Second heat exchanger 46 moderates the temperatures of Reactants A and B as the two reactants enter mixing chambers in a subsequent layer of the preferred reactor.

[0055] Fourth simple plate 40 incorporates important features that effect the fluid paths of Reactants A and Reactants B. Collectively, these features are referred to as an "inter-digital-mixer." The purpose of the inter-digital-mixer is to precisely align the fluid paths of Reactants A and B, such that a stacked laminar flow is enabled, while also ensuring that an equal pressure drop is achieved for the two reactants. A stacked laminar flow is preferred to enable diffusion mixing to occur. Diffusion mixing is recognized as being both extremely fast and efficient.

[0056] Reactant A flows through a series of openings 45, while Reactant B flows through a series of openings 47. Further details of the preferred stacked laminar flow will be provided below in conjunction with FIGURES 20-23. A series of dashed lines has been included on FIGURE 4, to illustrate that certain portions of openings 45 and 47 are aligned. As will become clear in examining the details of the fifth layer of the preferred reactor and a simple plate 50, as illustrated in FIGURE 5, once Reactants A and B have passed through the fluid paths formed by openings 45 and 47, the fluid paths of the reactants will be aligned. Simple plate 40 also includes temperature sensor opening 19.

[0057] Referring now to FIGURE 5, a fifth simple plate 50 includes chamfer 11. Heat transfer media A passes through

the fifth layer of the preferred reactor via heat transfer media A intake manifold 42a, and out through heat transfer media A exhaust manifold 42b. Fifth simple plate 50 also includes an opening corresponding to second heat exchanger 46. The purpose of having second heat exchanger 46 occupy two layers is to both increase the fluid capacity of second heat exchanger 46, and to also reduce the surface area of both simple plates 40 and 50, to provide better bonding. A functional reactor could be achieved by replacing second heat exchanger 46 in simple plate 40 with heat transfer media B intake manifold 26a and heat transfer media B exhaust manifold 26b (as in simple plates 20 and 30); however, the second heat exchanger of such a reactor would be slightly less effective.

[0058] The fifth layer of the preferred reactor also incorporates a plurality of fluid openings for Reactants A and B. Note that in the fifth layer, Reactants A and B have yet to be mixed together. The inter-digital-mixer of fourth layer (openings 45 and 47 in simple plate 40) has arranged the fluid paths of the reactants so that they are aligned in an alternating pattern. Notice that the dashed lines in simple plate 40 correspond to the plurality of Reactant A and B fluid openings in simple plate 50. The reactant fluid openings in simple plate 50 include twelve Reactant A openings 55, and twelve Reactant B openings 57. These openings are aligned in four rows of six openings, each row comprising three fluid openings for Reactant A, and three fluid openings for Reactant B, in an alternating pattern. Simple plate 50 also includes temperature sensor opening 19.

[0059] FIGURE 6 provides details of the sixth layer of the preferred reactor, showing a sixth simple plate 60 that includes chamfer 11. Heat transfer media A flows through the sixth layer using a heat transfer media A intake manifold 42a and a heat transfer media A exhaust manifold 42b. It is in the sixth layer that the Reactants A and B are first intermingled. The sixth layer incorporates four stacked laminar flow fluid channels 65, which are fed from the as yet un-mingled, alternating fluid openings 55 and 57 in fifth simple plate 50. Thus, fluid channels 65 convey both Reactant A and Reactant B. As will be described in detail below, the reactants flow through fluid channels 65 in a stacked laminar flow pattern. The sixth layer also includes temperature sensor opening 19.

[0060] Note that second heat exchanger 46 of fourth and fifth simple plates 40 and 50 is adjacent to a solid portion of simple plate 60. Thus second heat exchanger 46 is modifying the temperature of that solid portion of sixth simple plate 60, which in turn will modify the temperature of a corresponding portion of the next layer of the reactor.

[0061] The seventh layer of the preferred reactor includes a plurality of mixing chambers, which are fed from the combined fluid channels 65 of simple plate 60. FIGURE 7 shows a seventh simple plate 70 that also incorporates chamfer 11. Heat transfer media A flows through the seventh layer via a heat transfer media A intake manifold 42a and a heat transfer media A exhaust manifold 42b. Reactants A and B flow through four fluid channels 75 in a stacked laminar flow pattern. These fluid passages are fed from the corresponding fluid channels 65 in the sixth layer. Four fluid channels 75 lead to four mixing chambers 77, one mixing chamber being included for each fluid passage. These mixing chambers provide sufficient residence time for rapid diffusion mixing to occur.

[0062] It should be noted that seventh simple plate 70 does not incorporate an opening for a temperature sensor. Preferably a temperature sensor passes through the first six layers of the reactor and is disposed immediately above seventh simple plate 70. In this position, the temperature sensor is positioned to be able to monitor the temperature of the mixture of Reactants A and B in one mixing chamber 77. The temperature thus monitored is used to determine parameters for heat transfer media C (which services second heat exchanger 46 of the fourth and fifth layers). The optimal temperature range for reactants in most reactions is known. In some cases, the temperature sensor will indicate that Reactants A and B in mixing chamber 77 are not yet at the desired temperature. Then heat transfer media C will be used to increase the temperature of the reactants in the mixing chambers (via second heat exchanger 46). In other cases, the temperature sensor (not shown) will indicate that Reactants A and B in mixing chambers 77 are hotter than the desired temperature, and second heat exchanger 46 will be used to lower the temperature of the reactants. Note that heat exchanger 26 in the second layer is used to modify the temperature of Reactants A and B in the inter-digital-mixer of the fourth layer to a desired level. Generally, that desired level will be the reaction temperature. It is possible for heat exchanger 26 to be used to modify the temperatures of Reactants A and B to a temperature that is not the same as the reaction temperature, and then to use heat exchanger 46 to modify the temperatures of Reactants A and B to obtain the desired reaction temperature.

[0063] In the eighth layer of the preferred reactor, mixed Reactants A and B move from mixing chambers 77 and pass to lower layers of the reactor. FIGURE 8 shows an eighth simple plate 80 that also incorporates chamfer 11. Heat transfer media A passes through the eighth layer, using a heat transfer media A intake manifold 82a, and a heat transfer media A exhaust manifold 82b. It should be noted that the size of heat transfer media A intake and exhaust manifolds 82a and 82b have changed relative to the sizes of the heat transfer media A intake and exhaust manifolds 42a and 42b of the preceding layers. The purpose of this change in size will become clear in examining a ninth layer of the preferred reactor, as shown in FIGURE 9. Simple plate 80 includes a plurality of mixed reactant opening 85, which are in fluid communication with mixing chambers 77.

[0064] The ninth layer includes a third heat exchanger 93 that is used to moderate the temperature of the reactants as they pass through reaction channels in an eleventh layer of the preferred reactor. The ninth layer, as illustrated in FIGURE 9, is a ninth simple plate 90 having chamfer 11. Now, mixed Reactants A and B flow through ninth simple

plate 90 via mixed reactant openings 85. The path of heat transfer media A through ninth simple plate 90 is relatively complex, as compared to the paths of heat transfer media A through the previous layers of the preferred reactor. Heat transfer media A flows through ninth simple plate 90 via an intake manifold 42a, and an exhaust manifold 42b. Heat transfer media A is also flowing through third heat exchanger 93, which has a series of cutouts on both the right and left edges of the heat exchanger. These cutouts are disposed such that they overlap the enlarged areas of heat transfer media A intake and exhaust manifolds 82a and 82b of eighth simple plate 80. In this manner, heat transfer media A intake manifold 82a of eighth simple plate 80 services both heat transfer media A intake manifold 42a of ninth simple plate 90, and also heat exchanger 93 of ninth simple plate 90. Heat exchanger 93 is used to modify the temperature of a plurality of reaction channels in an eleventh layer, as will be described more in detail below, in conjunction with FIGURES 10 and 11.

[0065] It should be noted that the shape of third heat exchanger 93 has been designed and empirically tested to maximize fluid flow and heat transfer. For instance, the cut outs on the right side of third heat exchanger 93 are required for mixed reactant openings 85 to be included in layer 9. The left side of third heat exchanger 93 similarly has cut outs, but no mixed reactant openings 85 are located on the left side of simple plate 90, and theoretically cut outs are not required. However, as will be seen in FIGURE 13, a fourth heat exchanger is required to have cut outs on the left side for product openings to pass through layer 13. Because the third and fourth heat exchangers moderate the temperature of the same area, reaction channels in layer 11, the fluid dynamics of both heat exchangers should be as similar as possible. Thus, while each heat exchanger is required to have cut outs on only one of the right or left side for fluid openings, each heat exchanger has been designed with cut outs on both the right and left sides to achieve as much fluidic equilibrium as possible.

[0066] The indentation on the top edge of third heat exchanger 93, and a similarly shaped protrusion on the bottom edge of third heat exchanger 93, are included so that the flow of heat transfer media within third heat exchanger 93 matches as closely as possible the flows of reactants/product through reaction channels in layer 11. The shapes of the reaction channels and the third heat exchanger have been designed to enable other openings to exist on the simple plates (such as intake and exhaust manifolds, and other required fluid passages) and to match the fluid paths of the heat transfer media to the reactants fluid paths, as closely as possible. Compare the shape of third heat exchanger 93 to the reaction channels of layer 11, and the similarity will be apparent. If the upper and lower indentation and protrusion were not included in third heat exchanger 93, then the flow of heat transfer media through the third heat exchanger would generally flow from the cut outs on the right to the cut outs on the left, with little fluid flowing between these parallel flows. As a result, the heat transfer media in third heat exchanger 93 would have a flow pattern that does not match the flow pattern of mixed Reactants A and B in the reaction channels of layer 11, thus reducing the effectiveness of the third heat exchanger.

[0067] FIGURE 10 provides details of a tenth layer of the preferred reactor. FIGURE 10 shows a tenth simple plate 100 having chamfer 11. Heat transfer media A flows through the tenth layer using a heat transfer media A intake manifold 82a, and a heat transfer media A exhaust manifold 82b. Note again that the size of the intake and exhaust manifolds for heat transfer media A in the tenth layer have changed, now matching the size of the heat transfer media A intake and exhaust manifolds of the eighth layer. However, in the eighth layer, the purpose of the size change in the heat transfer media A intake and exhaust manifolds was to feed heat exchanger 93 in the ninth layer. The reason for the change in size of the heat exchanger intake and exhaust manifolds in the tenth layer is not related to servicing a heat exchanger. As noted above, the less surface area a simple plate has, the stronger the bond between the simple plates. The enlarged openings reduce the surface area, thus improving the bond strength. Another benefit is that, as will become apparent as later Figures are examined, tenth simple plate 100 (with the exception of the location of chamfer 11) is a mirror image of a subsequent simple plate 120 in the twelfth layer. Simple plate 120 is required to have the larger heat transfer media A intake and exhaust manifolds to feed a fourth heat exchanger in the thirteenth layer immediately below. Merely by changing the location of chamfer 11, the same fabrication configuration can be used to manufacture simple plate 100 and simple plate 120.

[0068] It should be noted that third heat exchanger 93 of the ninth layer of the preferred reactor modifies the temperature of a solid portion of tenth simple plate 100. As will be seen in examining an eleventh simple plate 110 of the eleventh layer of the preferred reactor, that solid portion corresponds to a plurality of reaction channels in eleventh simple plate 110. Because such a large portion of tenth simple plate 100 is required to be solid to provide for a heat transfer surface, the use of larger heat transfer media A intake and exhaust manifolds is important to reduce the surface area of simple plate 100, to increase the bond strength. The mixture of Reactants A and B flows through tenth simple plate 100 via a plurality of mixed reactant openings 85.

[0069] FIGURE 11 provides details on the eleventh layer of the preferred reactor, showing eleventh simple plate 110, with chamfer 11. Heat transfer media A flows through eleventh simple plate 110 via heat transfer media A intake manifold 42a, and heat transfer media A exhaust manifold 42b. The mixed Reactants A and B enter a plurality of reaction channels 115 from the right side of plate 110. Reaction channels 115 are fed from the plurality of mixed reactant openings 85, which form a passage from mixing chambers 77 of the seventh layer, through aligned mixed reactant openings 85

in simple plates 80, 90, and 100. The purpose of reaction channels 115 is to provide sufficient residence time within the preferred reactor so that the reaction between Reactants A and B can proceed either completely or primarily to completion. It is contemplated that should a particular reaction require addition time for completion, a separate residence chamber can be added in a layer downstream in the preferred reactor.

[0070] As was mentioned in relation to tenth simple plate 100 in FIGURE 10, the twelfth layer is required to have enlarged heat transfer media A intake and exhaust manifolds to feed a fourth heat exchanger in the thirteenth layer. Note that a twelfth simple plate 120 as shown in FIGURE 12 is quite similar to tenth simple plate 100. In fact, the layers are identical except for the location of chamfer 11. Thus, the same configuration pattern can be used to manufacture both tenth simple plate 100 and twelfth simple plate 120 other than location of the chamfer. Heat transfer media A flows through twelfth simple plate 120 via enlarged heat transfer media A intake manifold 82a and enlarged heat transfer media A exhaust manifold 82b. Twelfth simple plate 120 also includes a plurality of product openings 125, which are connected in fluid communication with reaction channels 115 of eleventh simple plate 110. These openings are identical in size to mixed reactant openings 85 of preceding layers, but product openings 125 are disposed on the left side of simple plate 120 rather than the right side of the simple plates (as the mixed reactants flow through the reaction channels from right to left).

[0071] The thirteenth layer of the preferred reactor incorporates a fourth heat exchanger, which is separated into two sections, an upper fourth heat exchanger 133a, and a lower fourth heat exchanger 133b. The fourth heat exchanger is also used to moderate the temperatures of the mixture of Reactants A and B flowing through reaction channels 115 of eleventh simple plate 110. FIGURE 13 shows a thirteenth simple plate 130 that includes chamfer 11. Reacted product from Reactants A and B passes through product fluid openings 125, into lower levels of the reactor.

[0072] As was the case in the tenth layer, the flow of heat transfer media A through thirteenth simple plate 130 is relatively complex. Heat transfer media A flows into a heat transfer media A intake manifold 42a and a heat transfer media A exhaust manifold 42b from similarly located (but larger) manifolds in twelfth simple plate 120. It should be noted that heat transfer media A intake manifold 42a and exhaust manifold 42b are not actually needed in thirteenth simple plate 130 to feed any heat exchanger in a subsequent layer. These manifolds are included to maintain the proper fluid pressure of heat transfer media A in the preferred reactor. If these manifolds were absent, the fluid pressure of heat transfer media A in third heat exchanger 93 of the ninth layer would be different than in the fourth heat exchanger (upper 133a and lower 133b), which is undesirable, as both the third and fourth heat exchangers are moderating the temperature of the reactants in reaction channels 115 of the eleventh layer, and thus both the third and fourth heat exchangers should have similar flow characteristics.

[0073] It should also be noted the incorporation of the heat transfer media A intake and exhaust manifolds in thirteenth simple plate 130 simplifies the production process. Note that thirteenth simple plate 130 is structurally very similar to the ninth simple plate 90 illustrated in FIGURE 9. With minor modifications, ninth simple plate 90 can be modified to yield thirteenth simple plate 130. As noted with respect to FIGURES 10 and 12, the differences would include the location of chamfers 11, as well as the additional change of splitting heat exchanger 93 (of FIGURE 9) layer into two heat exchangers 133a and 133b for thirteenth simple plate 130.

[0074] Heat transfer media A is required to flow through thirteenth simple plate 130 to service fourth heat exchangers 133a and 133b. Heat transfer media A is fed into these heat exchangers in the same manner as heat transfer media is fed into heat exchanger 93 from FIGURE 9, i.e. via cutouts that overlap the enlarged heat transfer media A intake and exhaust manifolds of the immediately preceding layer. Intake manifold 82a and exhaust manifold 82b of FIGURE 12 are enlarged such that heat transfer media A can enter and exit fourth heat exchangers 133a and 133b.

[0075] In FIGURE 13, the reason that the single heat exchanger 93 of FIGURE 9 has been separated into two heat exchangers is so that a plurality of temperature sensors can be incorporated into the thirteenth layer. Temperature sensor openings 139a, 139b, and 139c enable three temperature sensors (not shown) to be disposed adjacent to the thirteenth layer. Such temperature sensors will provide good temperature information relative to the temperature of the combined Reactants A and B flowing through the reaction channels of eleventh simple plate 110. Note that temperature sensors disposed in passages 139a-139c will actually measure the temperature of solid portions of twelfth simple plate 120 against which these passages abut. However, as discussed above, one preferable characteristic of the material from which the simple plates are fabricated is that the material be thermally conductive, at least with respect to layers that are transferring thermal energy to or from a fluid channel and a heat exchanger. Note that if such a layer is sufficiently thin, then the thermal conductivity of most materials is adequate. The temperature of the product of Reactants A and B flowing through reaction channels 115 in eleventh simple plate 110 will be very similar to the temperature of corresponding sections of twelfth simple plate 120. Because the temperature of the product in reaction channels 115 directly relates to the overall yield and quality of the product, three temperature sensors rather than one are employed in the preferred reactor.

[0076] Because the temperature of the reactants and the resulting product is so critical to yield and quality, the preferred reactor sandwiches reactant channels 115 of layer 11 between third heat exchanger 93 of layer 9 and fourth heat exchangers 133a and 133b of layer 13, to enable better temperature control. Third heat exchanger 93 actually

modify the temperature of a solid portion of tenth simple plate 100 that forms the upper surface of reaction channels 115. Fourth heat exchangers 133a and 133b moderate the temperature of a solid portion of twelfth simple plate 120 that forms the lower surface of reaction channels 115. It should be noted that the heat exchangers of the preferred reactor actually moderate the temperature of a solid portion of simple plates both above and below the opening that corresponds to the heat exchanger, that the purpose of the heat exchangers is to control the temperature of the reacting product in reaction channels 115. Note that fourth heat exchangers 133a and 133b are moderating the temperature of a solid portion of both simple plate 120 of layer 12 and a simple plate 140 of layer 14. While moderating a solid portion of simple plate 120 does effect the temperature of the product in reaction channels 115, the moderation of the solid portion of simple plate 140 serves no functional purpose. In the preferred reactor, the modification of the temperature of non-target portions of simple plates, such as simple plate 140, does not cause any problems. However, it is envisioned that in different stacked plate reactors, such non-target temperature changes could be undesirable. In such reactors, a simple plate that does not conduct thermal energy (i.e. whose thickness is sufficient to prevent heat transfer) could be used to isolate the heat exchangers to avoid non-target temperature changes.

**[0077]** The fourteenth layer of the preferred reactor is a relatively simple layer, involving only product fluid openings to withdraw reacted product from the reactor, and passages for temperature sensors as described above. FIGURE 14 illustrates fourteenth simple plate 140, which includes chamfer 11. Temperature sensor passages 139a, 139b, and 139c enable temperature sensors to be inserted into the reactor to monitor the temperature of reaction channels 115 as discussed above. Product openings 125 are used to direct the reacted product to lower levels of the reactor.

**[0078]** The fifteenth layer is also relatively simple. FIGURE 15 illustrates a fifteenth simple plate 150, having chamfer 11. The plurality of product openings 125 from the fourteenth layer are combined into a single product channel 155. Again, three temperature sensor openings 139a, 139b, and 139c are included to enable temperature sensors to be passed deeper into the core of the reactor.

**[0079]** The final layer of the preferred reactor is the sixteenth layer. FIGURE 16 illustrates a sixteenth simple plate 160, also having chamfer 11. The single product channel 155 of the fifteenth layer is reduced in area to a single product outlet port 165. Again, temperature sensor openings 139a-139c are available so that temperature sensors can be inserted deeper into the reactor. It should be noted that in the preferred reactor the thickness of both the top simple plate 10 and the bottom simple plate 16 are significantly greater than the thickness of the intermediate simple plates. The greater thickness provides both greater structural integrity, as well as helping to thermally isolate the inner layers of the reactor from the outside environment.

**[0080]** FIGURE 17 is an exploded isometric view of a preferred reactor 170 that includes the sixteen layers described in regard to FIGURES 1-16. Simple plates 10-160 are shown stacked in order so that the relative positions of each simple plate to each other may be examined. The preferred dimensional thickness of each simple plate is as follows:

Top simple plate 10	3.0 mm.
Second simple plate 20	0.3 mm.
Third simple plate 30	0.3 mm.
Fourth simple plate 40	0.3 mm.
Fifth simple plate 50	0.3 mm.
Sixth simple plate 60	0.3 mm.
Seventh simple plate 70	0.2 mm.
Eighth simple plate 80	0.3 mm.
Ninth simple plate 90	0.6 mm.
Tenth simple plate 100	0.3 mm.
Eleventh simple plate 110	0.2 mm.
Twelfth simple plate 120	0.3 mm.
Thirteenth simple plate 130	0.6 mm.
Fourteenth simple plate 140	0.3 mm.
Fifteenth simple plate 150	0.3 mm.
Sixteenth simple plate 160	3.0 mm.

**[0081]** Simple plates 10 and 160 (the top and bottom simple plates) are thicker than other plates to provide greater structural stability. Simple plates 20-60, 100, 120, 140 and 150 are much thinner, to enhance heat transfer. As will be discussed below, a thickness of 0.3 mm provides a reasonable heat transfer ability for a wide variety of materials. Simple plate 70 is thinner by 1/3 to ensure proper laminar flow within mixing chambers 75. Simple plate 11 is the same thickness as simple plate 70, to maintain fluidic equilibrium conditions in the reactor. Simple plates 90 and 130 are thicker than other plates to provide a larger mass of fluid in the heat exchangers 93, 133a and 133b. It should be noted

that the preferred plate thickness represent sheet metal thickness' that are commercially available, and that the ready availability of such materials lowers production costs.

[0082] For simple plates that include solid portions used to transfer thermal energy to or from heat exchangers, a preferred thickness is about 0.3 mm. As plate thickness increases, mechanical stability increases and heat transfer ability decreases. The 0.3 mm thickness provides good heat transfer characteristics without sacrificing mechanical stability. When graphs representing mechanical stability as a function of plate thickness (50 $\mu$ m-1 mm) and heat transfer ability as a function of plate thickness (50 $\mu$ m-1 mm) are combined, the curves representing each functional relationship intersect at approximately 0.3 mm. It should be noted that this optimum value of 0.3 mm is independent of the actual material selected (glass, metal, plastic, etc.). While the shape of the curves defining the functional relationships change when a different material is selected, the intersection of the curves at 0.3 mm remains relatively constant. Thus, 0.3 mm represents a simple plate thickness that provides for reasonable heat transfer ability without sacrificing structural integrity.

[0083] FIGURE 18A illustrates a fluid flow path of Reactant A, as it enters top simple plate 10 and proceeds through the sixth simple plate 60 of reactor 170. Reactant A enters through inlet 15 in top simple plate 10, proceeds to second simple plate 20 of the second layer, and enters Reactant A distributor 25. Reactant A then passes to third simple plate 30 of the third layer, passing through four Reactant A openings 35. In fourth simple plate 40 of the fourth layer, Reactant A passes through four Reactant A openings 45, which are part of the inter-digital-mixer. As discussed above, the purpose of the inter-digital-mixer is to precisely align the fluid flows for Reactants A and B to optimize mixing in later layers of the reactor. The purpose of Reactant A openings 45 is to precisely align a plurality of Reactant A fluid paths with a plurality of Reactant B fluid paths, so that a stacked laminar flow can be achieved with equilibrated pressure drops. It should be noted that first heat exchanger 24 is used to bring both Reactants A and B to the proper temperature in the inter-digital-mixer of the fourth layer.

[0084] In the fifth layer, fifth simple plate 50 incorporates a plurality of reactant A openings that are aligned with a plurality of Reactant B openings. These openings form an alternating pattern of 24 openings in four rows of six openings each (for a total of 12 Reactant A openings and 12 Reactant B openings). In the sixth layer, sixth simple plate 60 incorporates four fluid channels 65. It is in the four channels 65 that Reactants A and B first intermingle. Because of the pattern of fluid paths for Reactants A and B enabled by the inter-digital-mixer, Reactants A and B enter channels 65 in a stacked laminar flow pattern.

[0085] FIGURE 18B illustrates the fluid path that Reactant B takes in entering the first six layers of a reactor 170. Reactant B enters top simple plate 10 through opening 17, passes through the second layer an identical Reactant B openings 17 in second simple plate 20. In the third layer, Reactant B enters Reactant B distributor 37 in third simple plate 30. In the fourth layer, Reactant B enters four Reactant B openings 47 in fourth simple plate 40. As noted above, openings 47 (and 45) are collectively referred to as the inter-digital-mixer. After passing through the fourth layer, Reactant B flows into twelve openings 57 in fifth simple plate 50, of the fifth layer. Reactant B then proceeds to the four fluid channels 65 on sixth simple plate 60, where Reactants A and B are first co-mingled.

[0086] FIGURE 18C illustrates the combined flows of Reactants A and B after passing through the sixth layer and proceeding through layers 7-16 of reactor 170. Reactants A and B as combined (in a stacked laminar flow pattern) flow through four fluid channels 75 on seventh simple plate 70. Channels 75 lead to four mixing chambers 77. In mixing chambers 77, the stacked laminar flow is compressed, further enhancing rapid diffusion mixing. Second heat exchanger 46 is used to control the temperature of the reactants as they mix in mixing chambers 77. After Reactants A and B become thoroughly mixed in mixing chambers 77, the now mixed Reactants A and B flow through a plurality of mixed reactant openings 85 on eighth simple plate 80. The mixed reactants then flow through the ninth and tenth layers via identical mixed reactant openings 85 in simple plates 90 and 100, respectively. The mixed reactants then flow into reaction channels 115 on eleventh simple plate 110. Reaction channels 115 preferably provide sufficient residence time so that the majority (if not all) of the reaction is complete. If reaction channels 115 do not provide sufficient residence time, then an additional residence time chamber can be added downstream of reactor 170. As noted above, the quality and yield of the desired reaction is greatly affected by the ability to control temperature during the reaction process. The preferred reactor provides heat exchangers on simple plates 90 and 130 to precisely control the temperature within reaction channels 115. If additional residence time chambers are required, then control of the temperature in the additional residence time chambers is also highly desirable. After passing through reaction channels 115 in the eleventh layer, the resulting product passes through a plurality of product openings 125 in simple plates 120, 130, and 140 of layers 12, 13 and 14, respectively. The eight individual product streams represented by these product openings are then combined into a single product channel 155 on fifteenth simple plate 150, of layer 15. This single product exits the reactor via a product outlet 165 on sixteenth simple plate 160, in the bottom (sixteenth) layer of the reactor.

[0087] FIGURES 19A-19C illustrate the fluid paths for heat transfer media A, B, and C throughout the preferred reactor. FIGURE 19A illustrates the fluid path for heat transfer media B, which services first heat exchanger 24 in the second layer. Heat transfer media B flows into heat transfer media inlet 14a in top simple plate 10 and proceeds to heat exchanger 24 on second simple plate 20. Heat transfer media B passes through heat exchanger 24, and exits

heat exchanger 24 via outlet port 14b in top simple plate 10. The purpose of heat-exchanger 24 is to adjust the temperature of the solid section of portion of the third layer that is immediately above the inter-digital-mixer (openings 45 and 47) in fourth simple plate 40. In this manner, heat exchanger 24 is moderating the temperatures of Reactants A and B prior to the reactants being mixed together. It is contemplated that for the majority of reactions; it will be desirable for Reactants A and B to be of similar temperature. Those of ordinary skill in the art will readily understand, however, that there may be some reactions in which Reactant A and Reactant B will preferably be kept at separate temperatures. It is contemplated that a different stacked plate design using the same principles of the invention can be designed and fabricated to provide for a separate heat exchanger to individually modify the temperatures of Reactants A and B.

[0088] FIGURE 19B illustrates the fluid path that heat transfer media C takes through layers 1-4 of the preferred reactor. Heat transfer media C enters the reactor through inlet 16a in top simple plate 10 and then proceeds through heat transfer media C intake manifolds 26a on simple plates 20 and 30, in layers 2 and 3, respectively. Heat transfer media C then enters heat exchanger 46 on fourth simple plate 40 of layer 4 and exits heat exchanger 46 by utilizing heat transfer media C exhaust manifolds 26b of simple plates 30 and 20, in layers 3 and 2, respectively. Heat transfer media C then exits the reactor using outlet port 16b of top simple plate 10. The purpose of second heat exchanger 46 is to modify the temperature of the solid portion of sixth simple plate 60 that corresponds to the mixing chambers 77 of seventh simple plate 70. Because the mixing of chemicals often spontaneously generates heat, a great deal of heat can be generated as Reactants A and B are thoroughly mixed. Second heat exchanger 46 is thus able to cool Reactants A and B while in mixing chambers 77, so that the temperatures of the reactants do not exceed the ideal temperature for the desired reaction. Second heat exchanger 46 occupies both the fourth and fifth layers (simple plates 40 and 50), to increase the capacity of the heat exchanger.

[0089] FIGURE 19C illustrates the fluid path for heat transfer media A as it passes through the first thirteen layers of preferred reactor 170. Heat transfer media A enters the reactor at top simple plate 10 via intake port 12a. The heat transfer media A then passes through identical heat transfer media A intake manifolds 22a on simple plates 20 and 30 of layers 2 and 3 respectively. Heat transfer media A continues to pass through heat transfer media A intake manifolds in layers 4, 5, 6 and 7, via intake manifolds 42a. It should be noted that intake manifolds 42a differ in size and shape relative to the intake manifolds 22a of layers 2 and 3. The functional purpose of the size change is both reduce potential pressure drops within the fluid paths of the reactor, as well as to reduce the surface area of simple plates 40-70 to enhance bonding.

[0090] In layer 8, the shape of heat transfer media A intake manifold 82a changes once again. The purpose of the size change between the heat transfer media A intake manifolds in layers 7 and 8 is so that heat transfer media A can be fed into two separate sections of the layer 9. In a first heat transfer media A fluid path in layer 9, heat transfer media A flows into a heat transfer media A intake manifold 42a, and from there to heat transfer media A intake manifold 42a of tenth simple plate 100 in layer 10. From there, heat transfer media A flows to heat transfer media A intake manifold 42a in layer 11, an enlarged heat transfer media A intake manifold in layer 12, and then to heat transfer media A intake manifold 42a in layer 13.

[0091] In a second heat transfer media A fluid path in layer 9, fluid flows out of heat transfer media A intake manifold 82a of eighth simple plate 80 and into third heat exchanger 93 on ninth simple plate 90 of layer 9. As discussed above, the purpose of third heat exchanger 93 is to moderate the temperature of the solid portion of layer 10 immediately adjacent to reaction channels 115 in layer 11. Heat transfer media A exits heat exchanger 93 by returning to layer 8 via heat transfer media A exhaust manifold 82a, which is enlarged and overlaps the right end of third heat exchanger 93.

[0092] Simple plate 100 of layer 10 includes enlarged heat transfer media A intake manifold 82a (as well as exhaust manifold 82b). It should be noted that reaction channels 115 of layer 11 are not quite long enough to overlap the enlarged heat transfer media intake and exhaust manifolds 82a and 82b, thus no heat transfer media enters reaction channels 115. Here, the functional purpose of the size change of the intake and exhaust manifolds is to reduce the surface area of tenth simple plate 100, to enhance bonding, rather than to feed a heat exchanger (as in layer 8 and eighth simple plate 80).

[0093] Referring now to layer 11, note that again the size and shape of heat transfer media A intake manifold 42a has changed relative to the intake manifolds of layers 8 and 10. This size change relates to maintaining a calculated fluidic equilibrium throughout the micro reactor. However, it is contemplated that the overall effect of the size change is relatively minor, and that an effective micro reactor can be achieved without changing the size of the intake manifolds on layer 11.

[0094] In layer 12, the size and shape of heat transfer media A intake manifold 82a is again enlarged, to once again divert some heat transfer fluid A into a second fluid path that services fourth heat exchangers 133a and 133b of layer 13. Heat transfer media A also flows into a heat transfer media A intake manifold 42a in layer 13. The functional purpose of heat transfer media A intake manifold 42a of layer 13 is to ensure that the fluid pressure within fourth heat exchangers 133a and 133b matches the fluid pressure within third heat exchanger 93. Note both the third and fourth heat exchangers are moderating the temperature of reaction channels 115, and thus preferably both heat exchanges should have similar flow characteristics.



[0095] Heat transfer fluid A that has entered fourth heat exchangers 133a and 133b exits layer 13 via heat transfer media A exhaust manifold 42b in layer 12. From there, heat transfer media A moves successively through heat transfer media exhaust manifolds 42b in layer 11, 82b in layer 10, 42b in layer 9, 82b in layers 7-4 and 22b in layers 3-2. Heat transfer media A finally exits the reactor via outlet 12b in top simple plate 10.

[0096] Generally the heat transfer media used in the preferred reactor will be liquids, although it is envisioned that selected gases may also be beneficially employed. Fluidized solid heat transfer media (such as sand or silica) are known in the art, and might be used, though the dimensions involved in the fluid channels of the preferred reactor raise the concern that the solid heat transfer media could cause clogging of the heat transfer pathways.

[0097] The final Figures provide detail regarding the operation of the inter-digital-mixer of simple plate 40, in layer 4, illustrating how it enables a stacked laminar flow to be achieved. As noted above, a beneficial characteristic of laminar flow is that such a flow pattern enables diffusion mixing to occur. Diffusion mixing is extremely rapid, and an additional benefit is that no mechanical or electrically powered stirring or agitation means are required to mix the reactants, thus simplifying the reactor design.

[0098] FIGURE 20 illustrates side elevational views of layers 5-7, which implement the stacked laminar flow of the reactants. Isometric views of simple plates 50-70 are shown with dashed lines indicating where the cross-sectional views have been taken from. It should be noted that the structures being discussed in relation to FIGURE 20 (Reactant A openings 55, Reactant B openings 57, fluid channels 65, fluid channels 75, and mixing chambers 77) are present in sets of four, and the cross-sectional view illustrated could have been taken from three other portions of simple plates 50-70 as well. It should be understood that while the side elevational view illustrates only one set of the four structures (for example, one fluid channel 65), the preferred reactor includes four sets of the structures being discussed (i.e., four fluid channels 65).

[0099] Fifth simple plate 50 includes Reactant A openings 55, and Reactant B openings 57. As can be clearly seen in the isometric view of fifth simple plate 50, these openings are grouped in four rows of six. Each of the four rows has three Reactant A openings 55, and three Reactant B openings 57, in an alternating pattern. Thus, the preferred reactor includes 12 Reactant A openings 55, and 12 Reactant B openings 57, while in the side elevational view of FIGURE 20, only three of each are visible (one of the four rows). Each row of openings leads to a fluid channel 65 in sixth simple plate 60. Each fluid channel 65 is in fluid communication with a corresponding fluid channel 75 in seventh simple plate 70, and each fluid channel in turn leads to a mixing chamber 77, also in the seventh layer. Note that the openings forming fluid channels 65 and 75 are superimposed on each other, in effect forming a continuous volume. As the commingled Reactants A and B move from this continuous volume into mixing chambers 77, the reactants move from a relatively large volume into a relatively small volume. This reduction in volume is further heightened because seventh simple plate 70 is 2/3 the thickness of sixth simple plate 60 (0.2 mm vs. 0.3 mm in thickness). As will be described in further detail below, this reduction in thickness enhances the diffusion mixing. Due to the laminar flow the stacking of the plurality of reactant streams remains as it is after the thickness reduction. After such diffusion mixing in mixing chambers 77, the now mixed reactants exit mixing chambers 77 via the plurality of mixed reactant openings 85 in layer 8 (not shown here, see FIGURES 8 and 18C). A dashed line in FIGURE 20 indicates an area 185 that corresponds to the fluid path of the reactants through layers 5-7.

[0100] FIGURE 21 is an enlarged view of area 185 of FIGURE 20. FIGURE 21 illustrates how alternating openings 55 and 57 for Reactants A and B result in a "stacked" laminar flow. As can be clearly seen in FIGURES 18B and 18C, the reactants are flowing from left to right through layers 5-7. Reactant A enters the combined volume of fluid channels 65 and 75 first, via Reactant A opening 55a. Because Reactant A from opening 55a is the first fluid entering the combined volume of fluid channels 65 and 75, it moves to the bottom of the combined volume. A very short time later, Reactant B enters the combined volume of fluid channels 65 and 75, via Reactant B opening 57a. Because Reactant A from opening 55a has reached the bottom of the combined volume 65/75 first, Reactant B from opening 57a is "stacked" on top of Reactant A from opening 55a.

[0101] Reactant A from opening 55b flows into combined volume 65/75 next, and is "stacked" on top of Reactant B from opening 57b. Reactants A and B from openings 57b, 55c, and 57c enter combined volume 65/75 and are similarly stacked on top of one another. The result is that in each of the four combined fluid channels 67/75 of the preferred reactor, a six layered fluid stack is formed, with Reactant A on the bottom, then Reactant B immediately above, and so on. The resulting order of the stack, from top to bottom, is:

1 <sup>st</sup> Layer	Reactant B from opening 57c
2 <sup>nd</sup> Layer	Reactant A from opening 55c
3 <sup>rd</sup> Layer	Reactant B from opening 57b
4 <sup>th</sup> Layer	Reactant A from opening 55b
5 <sup>th</sup> Layer	Reactant B from opening 57a
6 <sup>th</sup> Layer	Reactant A from opening 55a



[0102] The height reduction of the combined volume of fluid channels 65/75 to mixing chambers 77 further enforces a stacked laminar flow. Preferably the channel height is reduced by two thirds in the mixing area respective to the lamination channel (fluid channels 65 and 75), resulting in a thickness of each individual fluid layer of less than 50  $\mu\text{m}$ . In this dimension, mixing by diffusion is enforced. In a laminar flow regime, fluids stay together as stacked layers when being forced into a narrower mixing area. To achieve the same volumetric flow rate in the mixing area as in the lamination channel, the mixing area is broader than the lamination channels (fluid channels 65/75), as can be seen in the isometric view of simple plate 70 (see also FIGURE 7).

[0103] It is critically important that when the reactants enter the lamination channels (fluid channels 65/75), they are stacked one on top of the other, rather than side by side. In the preferred reactor, the inter-digital-mixer of layer 4 (fourth simple plate 40) ensures that the plurality of reactant fluid streams are properly aligned so that when they enter the lamination channels (fluid channels 65/75) the reactant streams are properly stacked on top of one another. FIGURE 22 illustrates one design for an inter-digital-mixer that *will not* ensure the reactants will properly stack on one another, while FIGURE 23 illustrates the preferred design of the inter-digital-mixer that *does* enable proper stacking.

[0104] In FIGURE 22, both Reactant A opening 45a and Reactant B opening 47 are similar in size and shape. These openings are shown as superimposed over fluid channel 65. It should be understood that openings 45a and 47 are part of fourth simple plate 40 (the fourth layer), while fluid channel 65 is part of sixth simple plate 60. However, openings 45a and 47 are in fluid communication, and are shown here together to illustrate the fluid path enabled by this poorly designed inter-digital-mixer. In FIGURE 22, an area 186 is indicated by dashed lines. In an enlarged view of area 186, details of the fluid paths enabled by this poorly designed inter-digital-mixer can be seen.

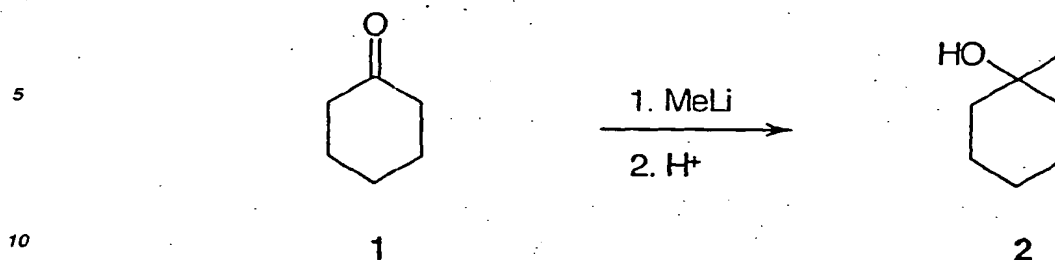
[0105] As noted above, the function of the inter-digital-mixer is to separate two individual reactant fluid paths into a plurality of fluid paths. It is important that these plurality of fluid paths are properly aligned, so that the stacked laminar flow described above can be achieved. Thus, the design of the inter-digital-mixer is extremely important, as a poorly designed inter-digital-mixer will not ensure that the desired stacked laminar flow is achieved.

[0106] In the enlarged detail of area 186, a Reactant A fluid path 145a and a Reactant B fluid path 147 are shown. Note that as illustrated, both the reactants enter fluid channel 65 in a side by side laminar flow pattern, rather than in the desired stacked laminar flow pattern, because the reactants are entering fluid channel 65 from opposing sides, rather than from the same side. The side by side effect is due to how the fluid fronts of each fluid path propagate. Note that each fluid path (145a and 147) is changing direction by approximately 90 degrees as the fluid path enters fluid channel 65. The fluid front of each fluid path will have a tendency to propagate fastest along the inside of that 90 degree corner, as the fluid on the inside of the corner has less distance to travel. When fluid path 145a encounters fluid path 147, each respective fluid front is maximized along the channel wall corresponding to the inside of the 90 degree corner associated with each fluid path. Thus a side by side laminar flow condition results.

[0107] Compare the poorly designed inter-digital-mixer of FIGURE 22 with the preferred inter-digital-mixer of FIGURE 23. The shape of Reactant A opening 45 has been changed to include a curve at the portion of the openings that are superimposed over fluid channel 65. These curves ensure that even during low pressure or low flow conditions, Reactant A enters fluid channel 65 from the same side as Reactant B. FIGURE 23 similarly includes dashed lines indicating an area 187. In the enlarged view of area 187, a Reactant A fluid flow 245 can be seen with a Reactant B fluid flow 247 stacked on top, rather than side by side, as in FIGURE 22. As noted above, in layers 4-7 (the inter-digital-mixer, fluid channels 65 and 75, and mixing chambers 77) the reactants flow from left to right, Reactant A enters fluid channel 65 first, and is thus on the bottom of the channel, with Reactant B stacked on top. Because the openings alternate between Reactant A and Reactant B, a six layer stacked laminar flow is achieved, with three layers of Reactant A and three layers of Reactant B in an alternating pattern. While additional simple plates could have been added to the preferred reactor to define a reactant fluid path that also ensured that the reactants would enter fluid channel 65 from the same side, this modification would not only increase the cost and complexity of the reactor, but it would also result in different pressure drops between the reactants. This difference would result in less than ideal mixing dynamics, and thus, is not a preferred solution. It should be noted that the shape of openings 45, in particular, the curve radius, length, and width have been carefully selected to achieve an equal pressure drop for both Reactants A and B.

#### Exemplary Chemical Reaction Performed in a Stacked Simple Plate Reactor

[0108] The described chemical reaction belongs to the class of organometallic conversions, i. e., the addition of an organolithium compound to a carbonyl compound. Cyclohexanone (1) reacts in a one step procedure with methyl lithium to produce the 1,2-addition product 1-methyl-cyclohexanol (2).



15 Supply of the Starting Materials:

[0109]

1. A 1.5 molar solution of methyl lithium dissolved in diethylether (commercially available in 100-ml bottles sealed with a septum).  
 2. Preparation of 100 ml of a solution of 13.2 grams (0.15 moles) of cyclohexanone (commercially available liquid) in dry diethylether.

[0110] Solution No. 2 is transferred into a pressure compensated bottle with tube connectors. Both solutions are connected to an argon atmosphere prior to use to avoid hydrolysis with air.

25 Thermal Conditioning and Setting up of the System:

[0111] The reactor temperature is adjusted to -20 °C by cryostats, which are connected to the heat exchangers of the reactor. Solvent (diethylether) is pumped continuously through the complete system until the solvent flow leaving the reactor has reached -20 °C.

30 Performing the Reaction:

[0112] After reaching thermal equilibrium, the two-reactant solutions are transferred by individual pumps via Teflon™ tubing into the reactor. The pump rate is set to 1 ml/min for each reactant. The two reactant flows are each divided into four parallel laminar flow streams with dimensions of several micrometers. They enter the inter-digital-mixer located under heat exchanger 1, which is a cross flow type heat exchanger, where they are cooled to the appropriate temperature (-20 °C). It should be noted that this temperature is a function of the desired reaction. In some cases, each reactant may require pre-treatment to obtain a different reactant temperature. Thus, the stacked simple plate reactor may preferably include a heat exchanger for modifying the temperature of each reactant. Of course, if the reactant temperatures are to be the same, then a stacked simple plate reactor can be designed with a single heat exchanger to pretreat both reactants.

[0113] In the inter-digital-mixer the two reactant flows are each divided again into 12 individual laminar flow streams for each reactant. These 24 streams enter the four lamination channels in groups of six streams, where each group of six streams are combined again (stacked onto each other). From here, the four groups of six stacked fluid streams enter the four mixing chambers (which are placed under heat exchanger 2), and the four groups of six stacked fluid streams are reduced in thickness so that diffusion mixing will occur. Thus in each of the four groups of six stacked fluid streams, three single streams of reactant A are united with three single streams of reactant B. After mixing, the reactants exit the four mixing chambers at two exits per mixing chamber, thus resulting in eight mixed reactant streams. These eight mixed reactant streams enter the eight reaction channels, which are sandwiched between heat exchangers 3 and 4. It is within the reaction channels that the final reaction takes place, and it is here that the most heat is generally produced; thus the reaction channels are sandwiched between two heat exchangers.

[0114] Heat transfer is extremely efficient due to the high surface to volume ratio, to the selection of an extremely thermally transparent material for the simple plates that form the heat exchanger (by the control of the material and/or the thickness of the simple plates), and to the thinness of the simple plates (the distance between the mixing zone and heat exchanger is in the range of a few micrometers). Thus, the heat of the exothermic reaction can be reduced to 1-2 °K above the determined reaction temperature.

[0115] The internal volume of the mixing zone is approximately 1 ml, providing a residence time of 30 seconds, during which the majority of the reaction is completed. For reactions that need a longer reaction time, an additional residence

time chamber can be added to the reactor, either by using additional simple plates, or by adding a separate residence chamber module downstream of the reactor.

[0116] The resultant product stream leaves the reactor via a Teflon™ tube into a collection flask that is filled with 2N hydrochloric acid. Instant quenching of the addition adduct and excess reagent takes place.

#### Benefits of the Simple Plate Stacked Reactor:

[0117] Advantages of the stacked simple plate reactor system are precise temperature control, exact adjustment of reaction time, and eliminating the need of a protective atmosphere, since the reactor is a closed environment. Enhanced safety is provided due to the small quantities of material, and the closed environment operating conditions.

[0118] The system is especially advantageous when large quantities of product are required, because the reactor can work continuously, and can be operated for hours, even up to days, without maintenance. Accordingly, automated production of large amounts of the desired product without the loss of efficiency and safety can be achieved. Additional product can be obtained by operating additional reactors in parallel under identical operating conditions.

#### System Description:

[0119] The reactants are provided in conventional laboratory bottles with tube connectors. The bottles are connected to a pump module by Teflon™ tubes. Inside a pump module disposed upstream from the pumps are three way valves, which are connected to the reactants, the solvents and pump inlet. For conditioning the stacked simple plate reactor, the valves are set to the solvents, so that the pumps first fill the whole system with solvent until the stacked simple plate reactor reaches thermal equilibrium. Then the valves are set to the reactants, enabling the pumps to deliver the reactants into the stacked simple plate reactor. A filter is placed inline between the pump outlet and reactor inlet to avoid clogging of the system by particulates. Fluidic connection of pumps and reactor can be achieved by commercially available HPLC fittings. Controlling the temperature of the stacked simple plate reactor is achieved by pumping heat transfer media from a cryostat into the internal heat exchangers of the stacked simple plate reactor. Product coming out of the system is collected in a conventional laboratory bottle.

#### Measuring and Automation Control Devices:

[0120] All pumps, valves and cryostats are preferably controlled by a microcontroller or computer, programmed with appropriate software, enabling convenient adjustment and control of the system. The following sensor devices are optionally used to provide analog signals that are converted to digital signals for input to the microcontroller or computer, to facilitate more efficient manual or automated control of the chemical process:

- Pressure sensors disposed downstream from each pump and at the inlet and outlets of the stacked simple plate reactor.
- Temperature sensors integrated in the stacked simple plate reactor and disposed close to the mixing zone and at the reactor outlet.
- Optional flow sensors introduced into each reactant stream for improved flow adjustment.

Excellent control and adjustment of flow and ratio of the reactants, determination of the pressure buildup inside the system by differential pressure measurement, and exact adjustment and control of the reaction temperature can thus be achieved.

[0121] Although the present invention has been described in connection with the preferred form of practicing it, those of ordinary skill in the art will understand that many modifications can be made thereto within the scope of the claims that follow. Accordingly, it is not intended that the scope of the invention in any way be limited by the above description, but instead be determined entirely by reference to the claims that follow.

#### **Claims**

1. Reactor for contacting and optionally reacting one chemical with at least one other chemical to form a chemical product, said reactor comprising a plurality of simple plates stacked in layers, each simple plate having at least one opening that extends therethrough, an opening in each simple plate overlapping at least one other opening in an adjacent simple plate, thereby forming at least one passage within the reactor to convey and mix said one chemical with said at least one other chemical, the chemical product being formed within said passage by a reaction between said one chemical and said at least one other chemical.

2. A reactor for carrying out a chemical process, comprising a plurality of simple plates stacked together in layers, said layers including at least one inlet port and at least one outlet port for the receipt and discharge of chemicals, respectively, and at least one pathway for accommodating the chemicals while they are processed, wherein said at least one pathway is connected in fluid communication with said inlet and outlet ports and comprises an opening through at least one simple plate aligned with an opening through an adjacent simple plate.

3. A chemical reactor according to claim 1 or 2 for processing at least two reactants to form a desired chemical product, comprising:

- (a) a stack of simple plates, including two outer simple plates and at least one intermediate simple plate;
- (b) inlet openings for receiving each of the reactants and an outlet opening for discharging the chemical product formed in at least one of the outer simple plates; and
- (c) an opening through each intermediate simple plate forming a reactant pathway, said reactant pathway being in fluid communication with each inlet opening, said at least two reactants mixing in said reactant pathway and reacting therein to produce the chemical product, said reactant pathway also being in fluid communication with the outlet opening, so that said chemical product is discharged from the chemical reactor through the outlet opening.

4. The chemical reactor of any of the preceding Claims, further comprising:

- (a) a first heat exchange fluid inlet port and a first heat exchange fluid outlet port through which a first heat transfer fluid is introduced into and discharged from the chemical reactor, said first heat exchange fluid inlet port and said first heat exchange fluid outlet port being disposed in at least one of the simple plates; and
- (b) a first heat exchanger defined by an opening in at least one intermediate simple plate in the stack and by adjacent simple plates disposed on opposite sides of said opening, said opening being in fluid communication with the first heat exchange fluid inlet port and the first heat exchange fluid outlet port.

5. The chemical reactor of any of the preceding Claims, further comprising:

- (a) a second heat exchange fluid inlet port and a second heat exchange fluid outlet port through which a second heat transfer fluid is introduced into and discharged from the chemical reactor, said second heat exchange fluid inlet port and said second heat exchange fluid outlet port being disposed in at least one of the simple plates; and
- (b) a second heat exchanger defined by an opening in at least one intermediate simple plate in the stack and by adjacent simple plates disposed on opposite sides of said opening, said opening being in fluid communication with the second heat exchange fluid inlet port and the second heat exchange fluid outlet port.

6. The chemical reactor of any of the preceding Claims, wherein the second heat exchanger is used to modify a temperature of at least one of the at least two reactants such that said at least one of the at least two reactants has a different temperature.

7. The chemical reactor of any of the preceding Claims, wherein at least one intermediate simple plate that includes openings defining the first fluid path and the second fluid path comprise an inter-digital-mixer that separates and aligns said first fluid path and said second fluid path, forming a plurality of individual fluid paths.

8. The chemical reactor of any of the preceding Claims, wherein the plurality of individual fluid paths are joined in a laminar flow pathway to provide a stacked laminar flow of said first and said second at least two reactants.

9. The chemical reactor of Claim 8, wherein a height of said laminar flow pathway is reduced to enhance the stacked laminar flow, preferably so that a flow rate of a fluid in said laminar flow pathway remains constant.

10. The chemical reactor of any of the preceding Claims, wherein at least one inter-digital-mixer maintains a substantially equivalent pressure drop for each of the individual fluid paths when said first fluid path and said second fluid path are separated and aligned into the plurality of individual fluid paths.

11. The chemical reactor of Claim 1 wherein an opening in a simple plate serves at least one of the following functions:

- (a) reduces a surface area of a simple plate, thereby enhancing bonding between adjacent simple plates;

(b) provides a fluid path for one of:

- i. said at least two reactants;
- ii. the chemical product; and
- iii. a heat transfer fluid;

(c) facilitates mounting a temperature sensor;

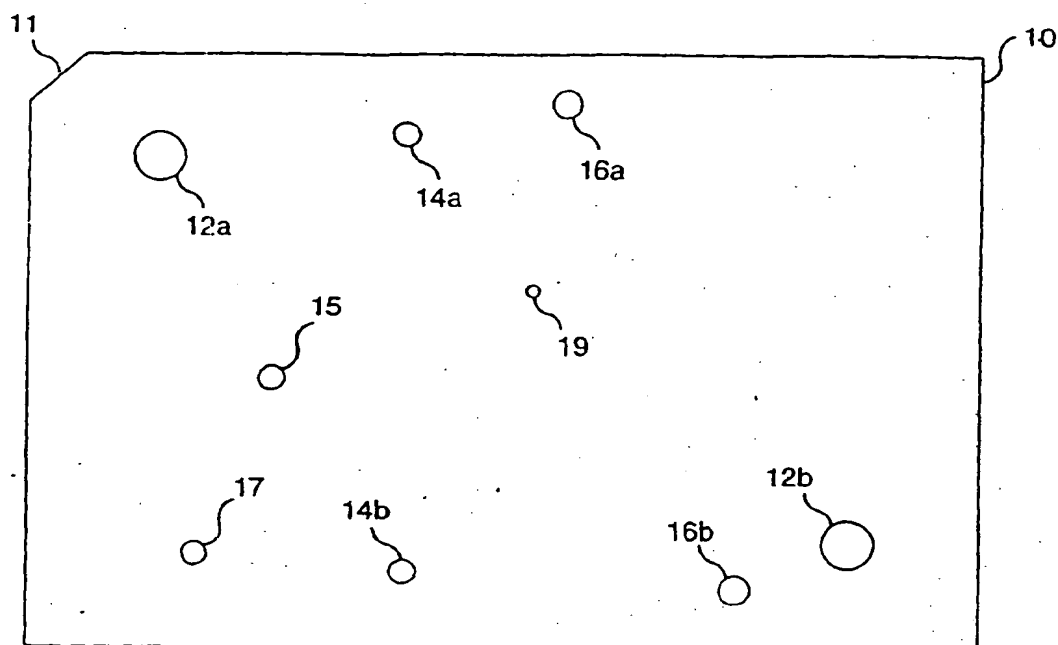
(d) separates a single fluid path into a plurality of fluid paths;

(e) provides at least one of a heat exchanger, a mixing chamber, an inter-digital-mixer, and a reaction pathway; and

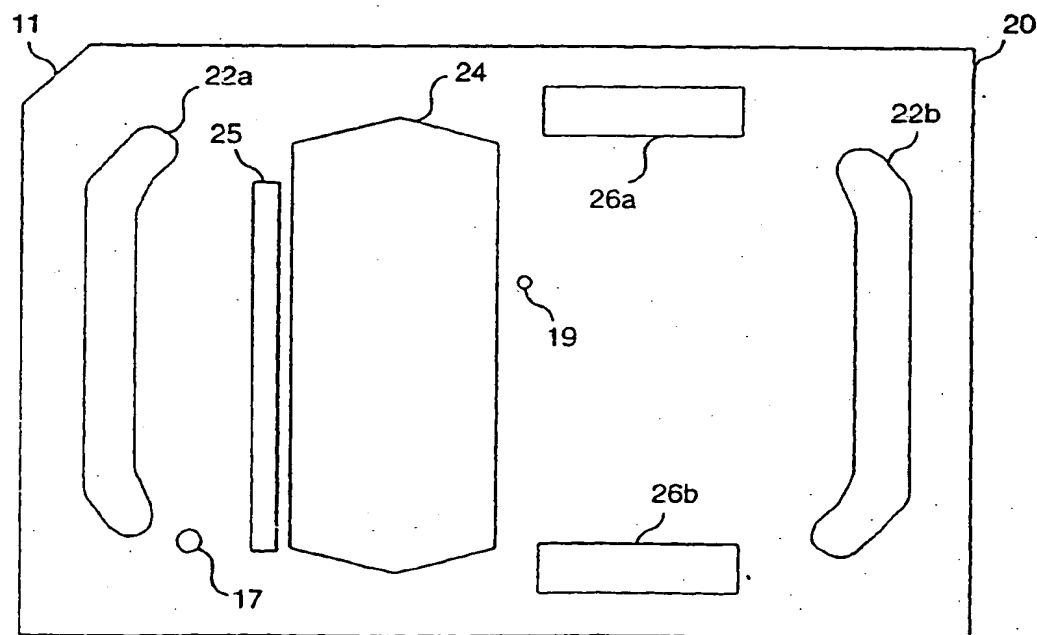
(f) enhances a flow characteristic of a fluid, including a direction of a fluid flow and a pressure drop of the fluid within the chemical reactor.

12. A method for fabricating a chemical reactor for combining at least two reactants to produce a desired product, comprising the steps of:

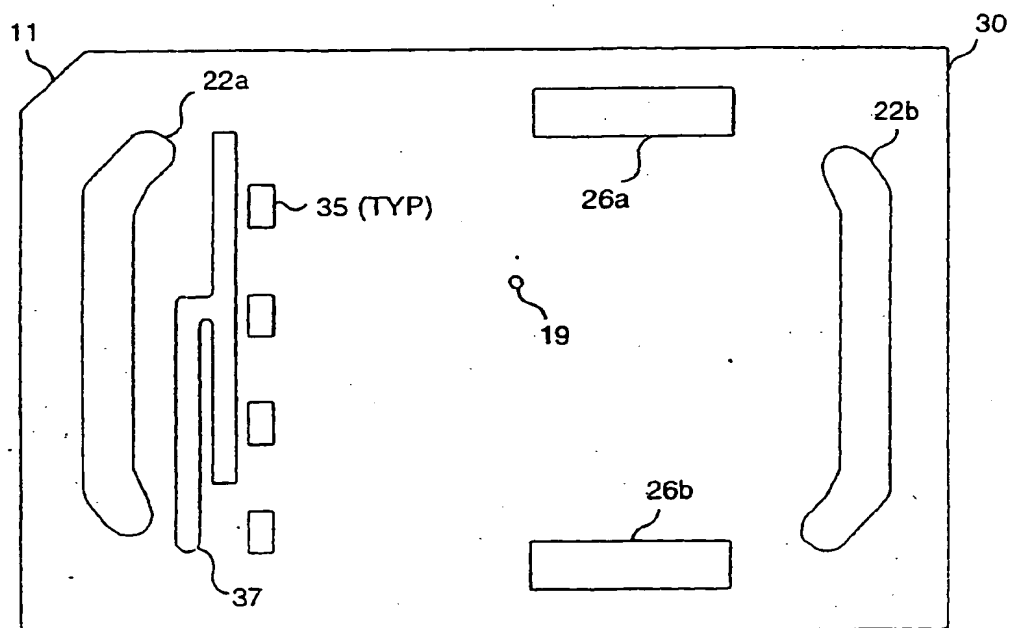
- (a) providing a plurality of simple plates, said plurality of simple plates including two outer simple plates and at least one intermediate simple plate in which is formed at least one opening, at least one of the outer simple plates including inlet ports for said at least two reactants and an outlet port for discharging the desired product;
- (b) stacking said plurality of simple plates such that said at least one opening in said at least one intermediate simple plate is in fluid communication the inlet ports and the outlet port; and
- (c) securing said plurality of simple plates in the stack.



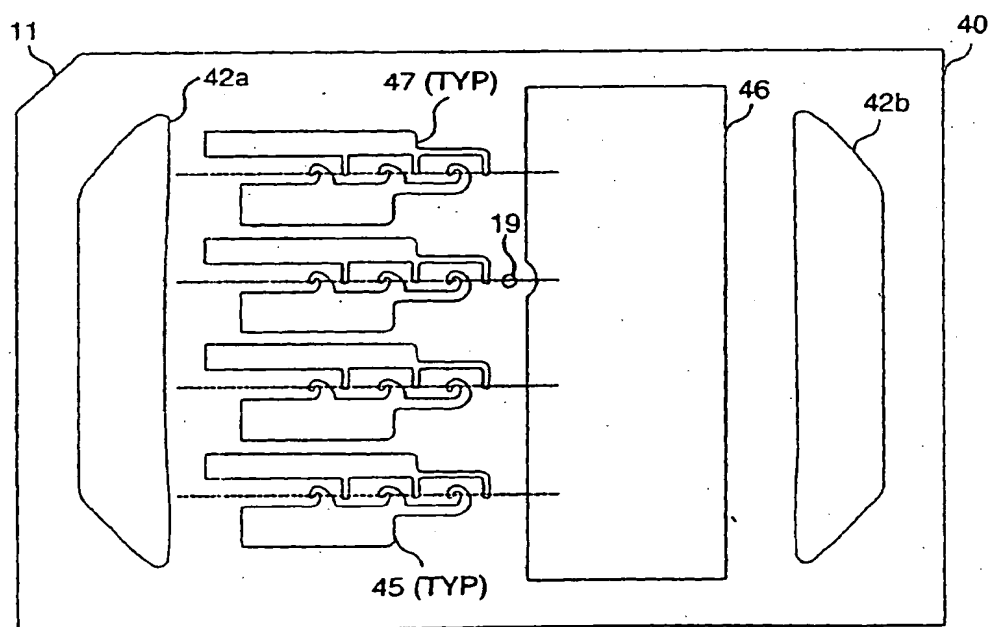
**FIG. 1**



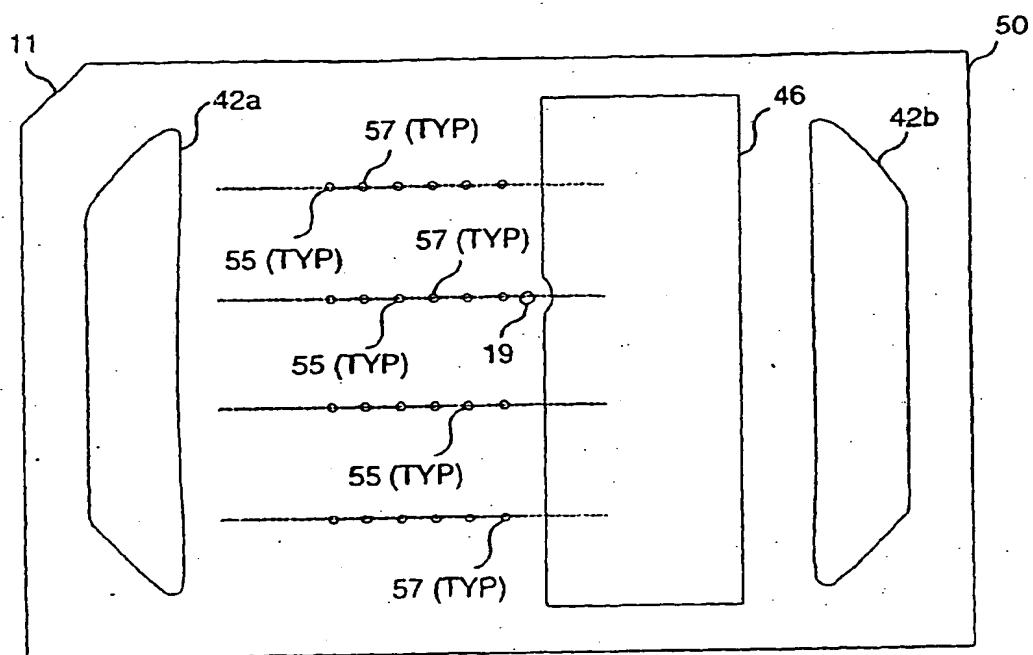
**FIG. 2**



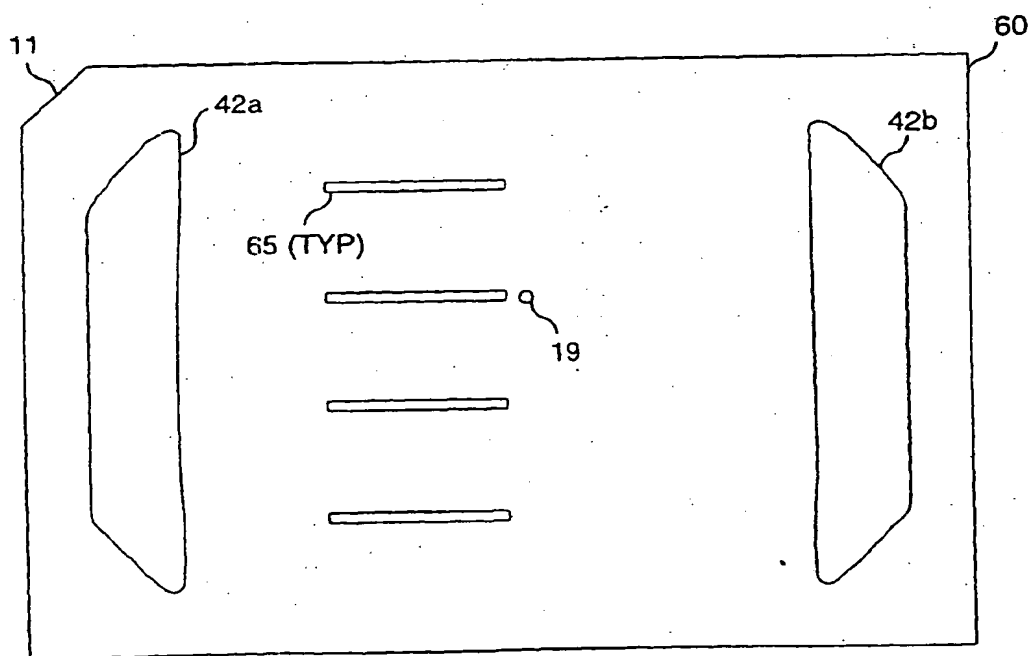
**FIG. 3**



**FIG. 4**

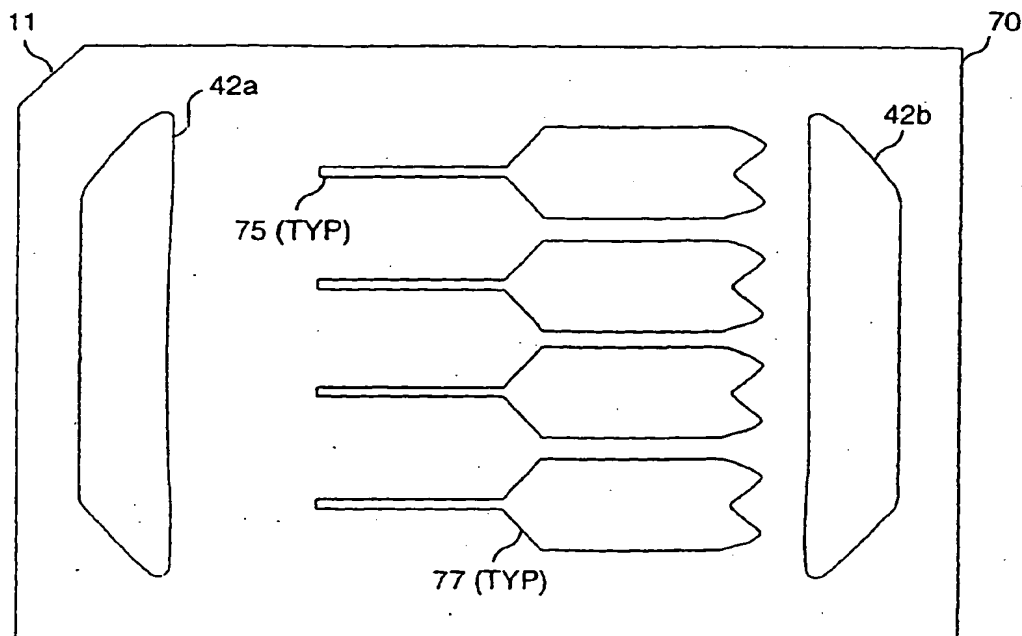


**FIG. 5**

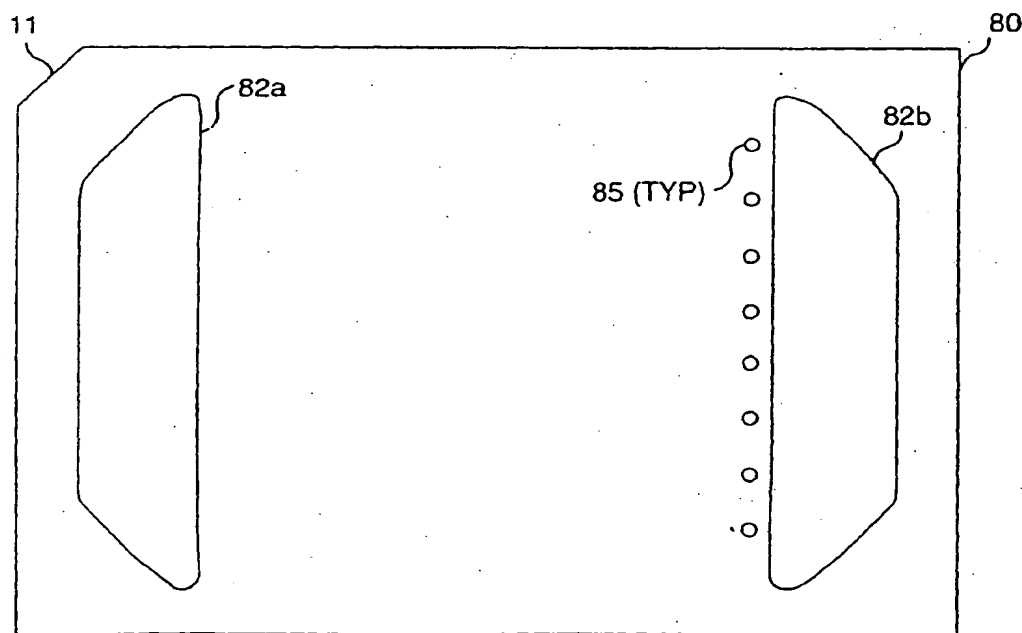


**FIG. 6**

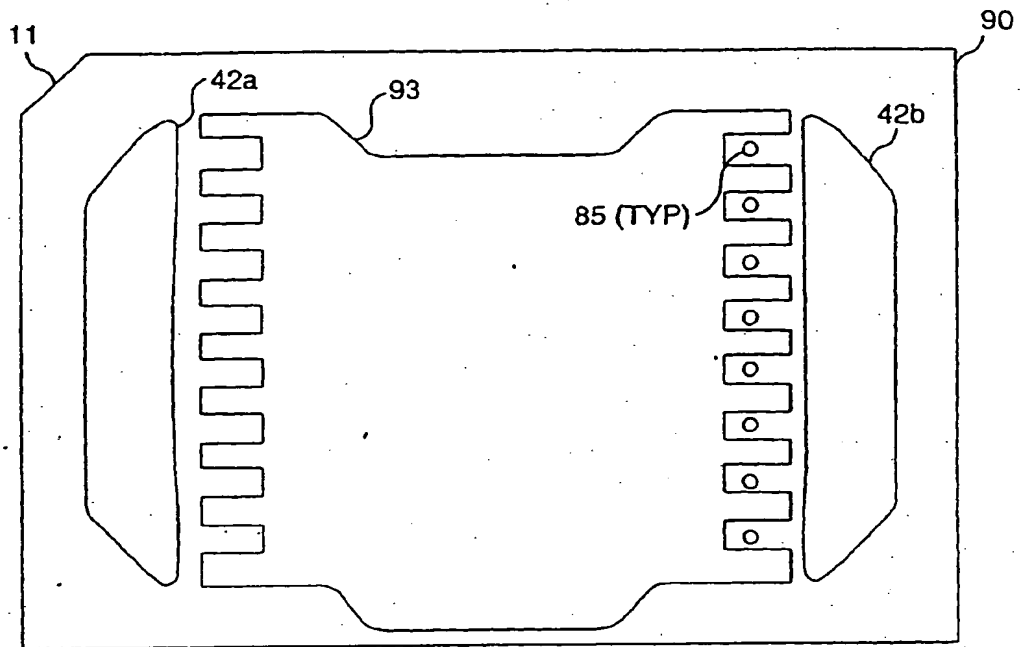




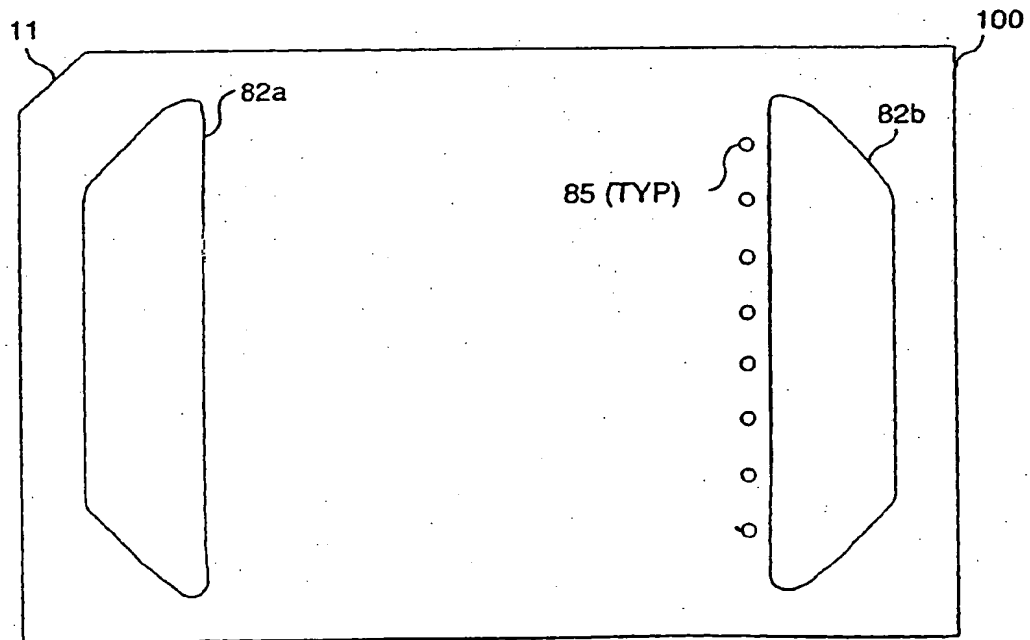
**FIG. 7**



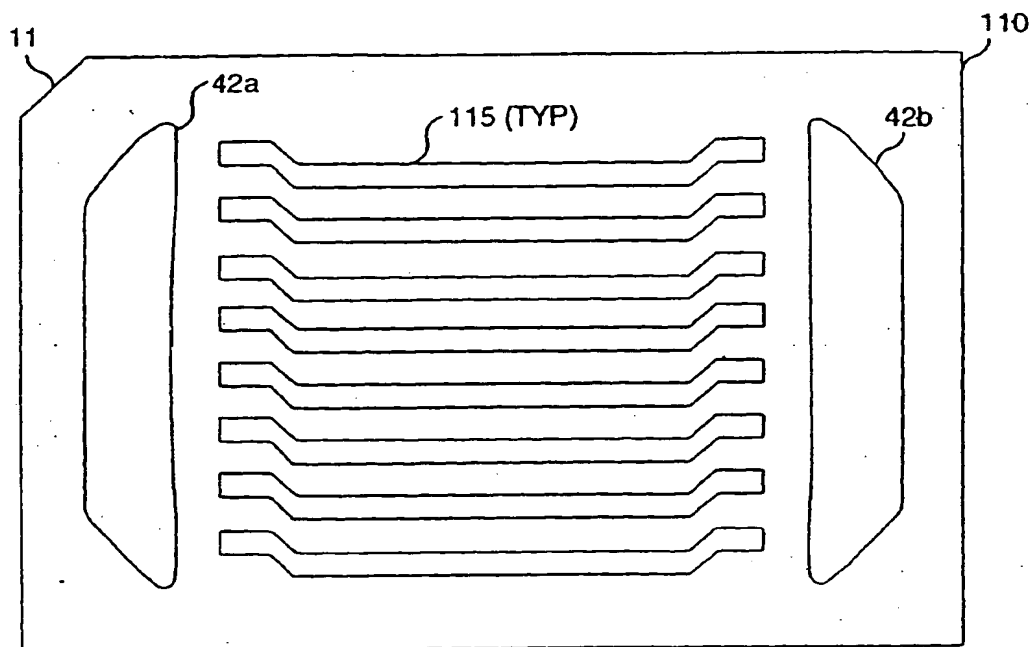
**FIG. 8**



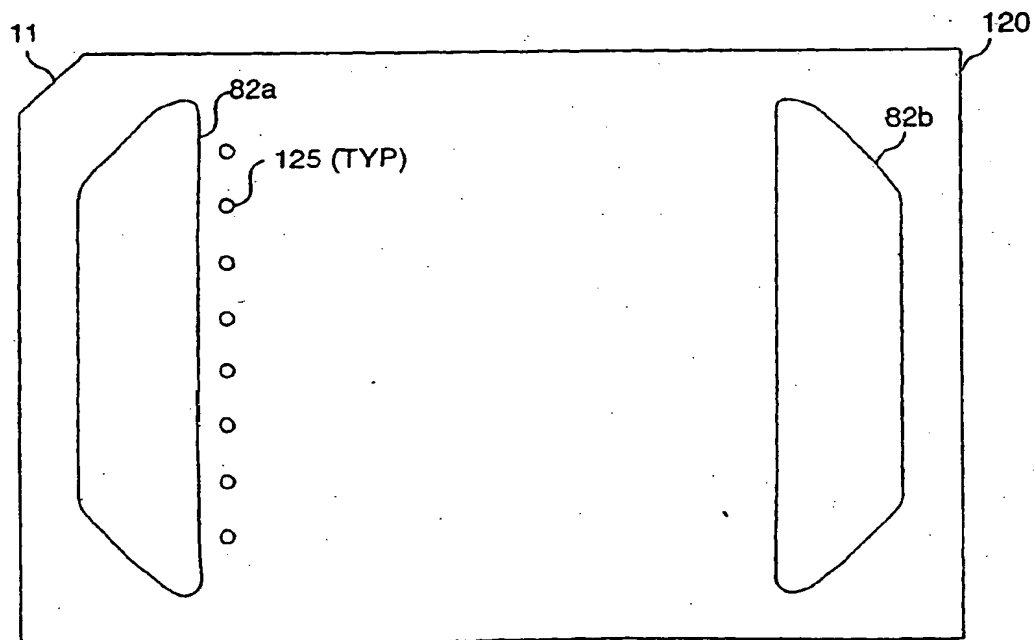
**FIG. 9**



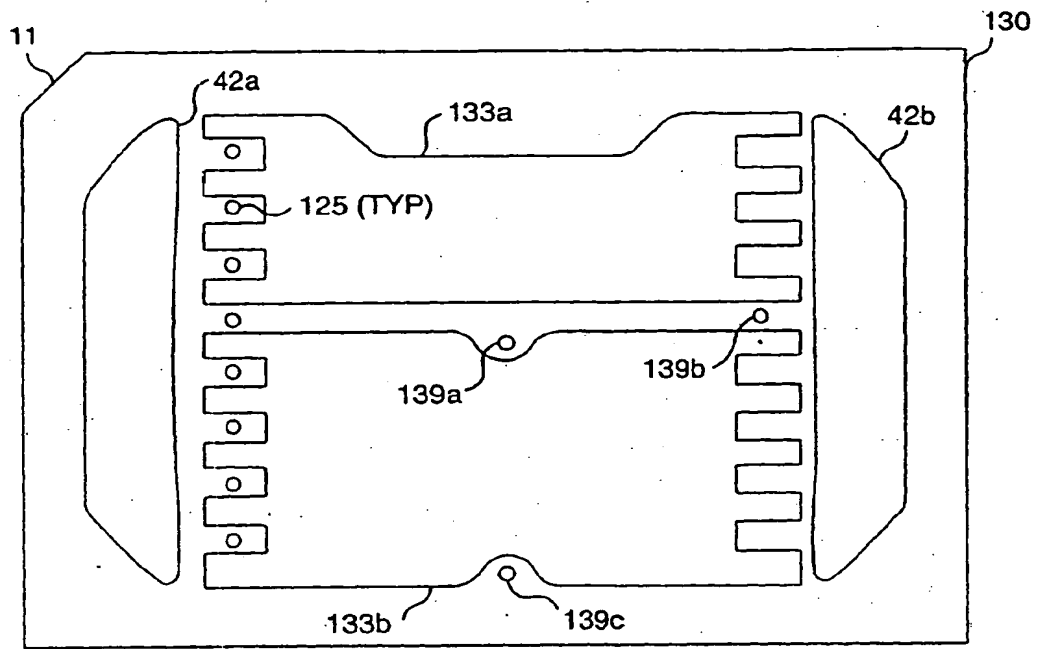
**FIG. 10**



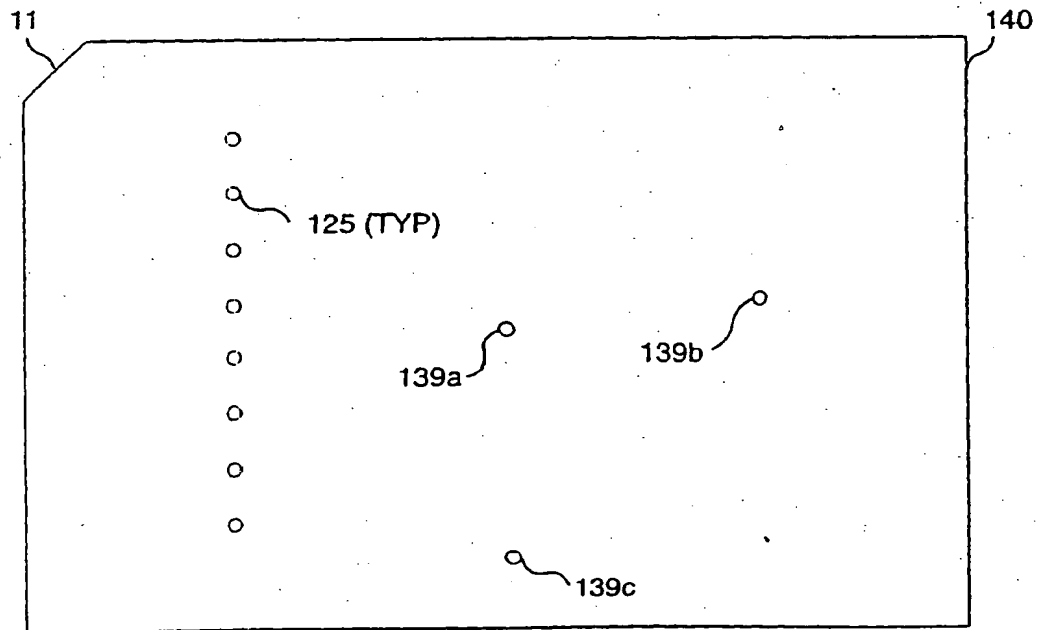
**FIG. 11**



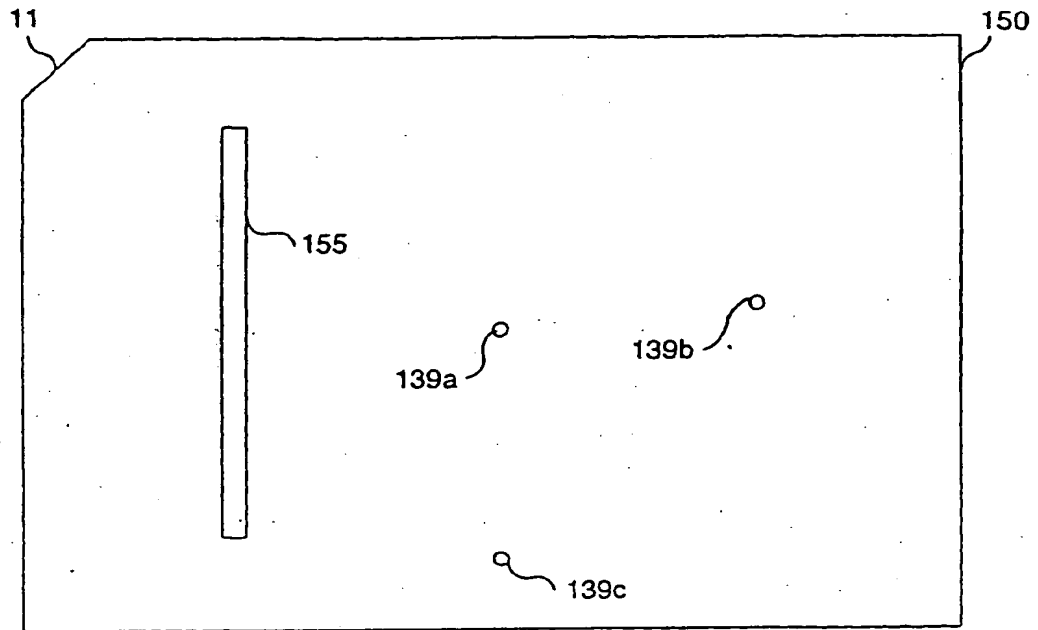
**FIG. 12**



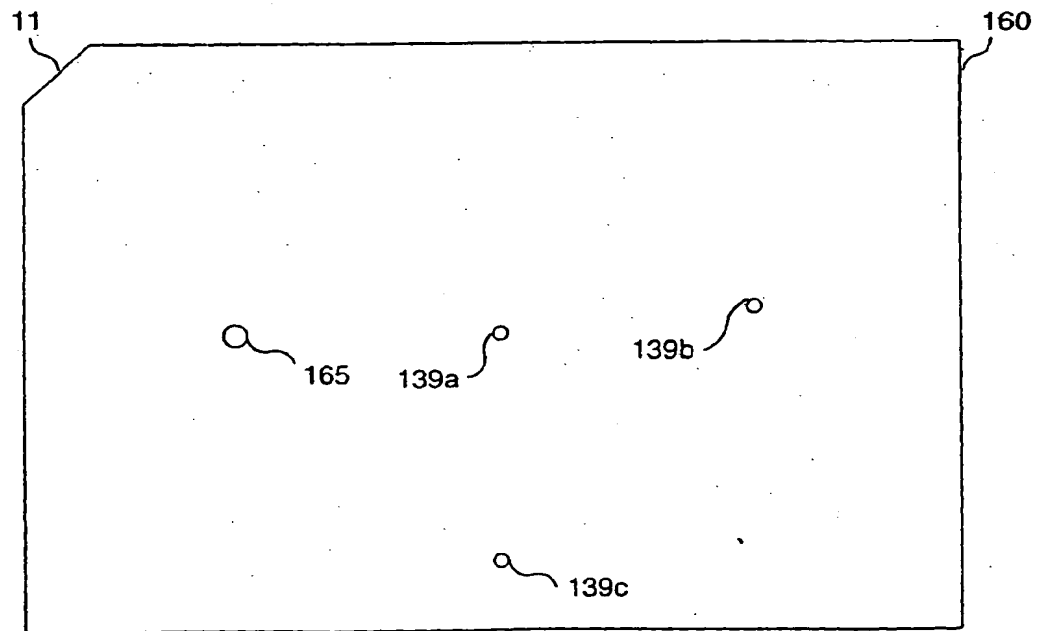
**FIG. 13**



**FIG. 14**

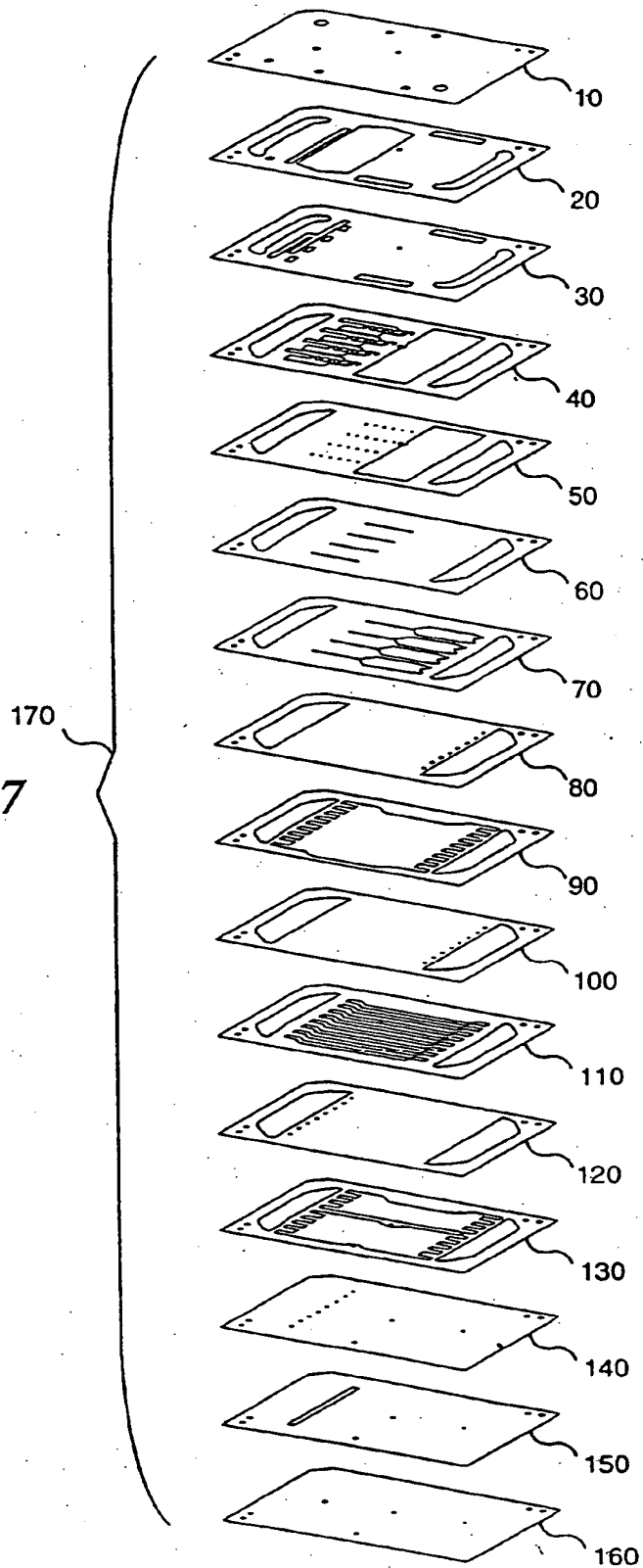


**FIG. 15**



**FIG. 16**

**FIG. 17**



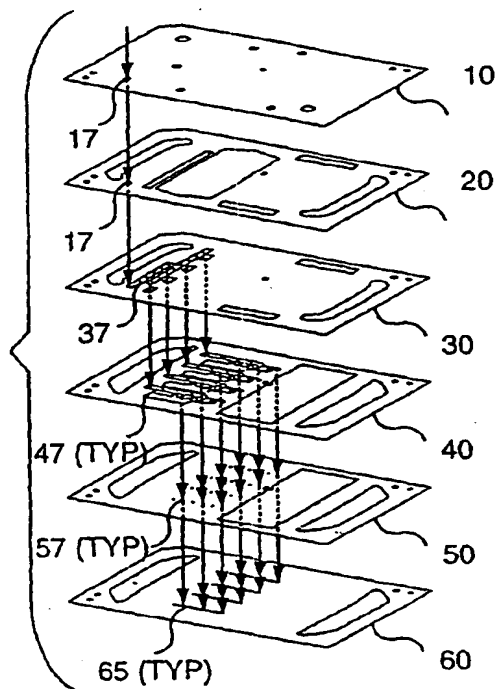
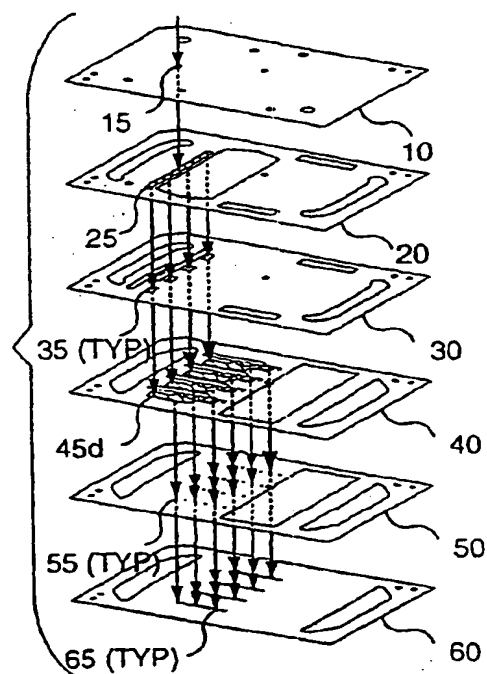


FIG. 18A



**FIG. 18B**

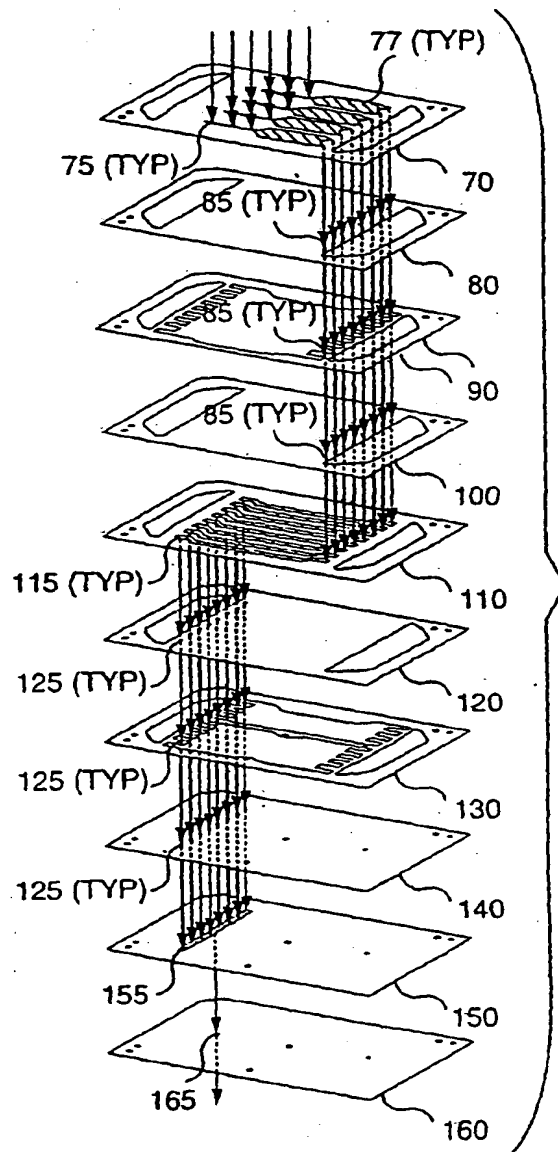


FIG. 18C

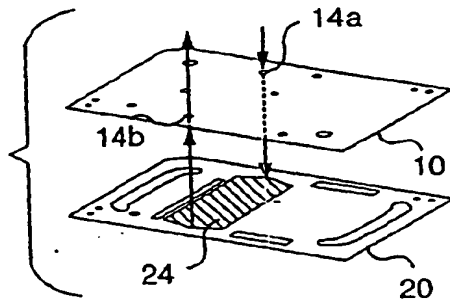


FIG. 19A

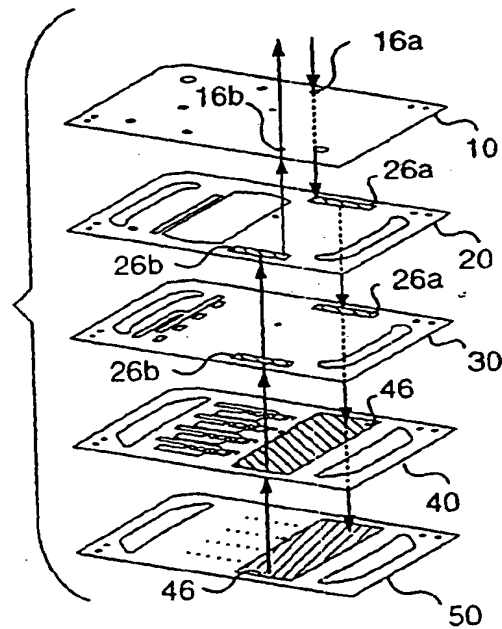


FIG. 19B

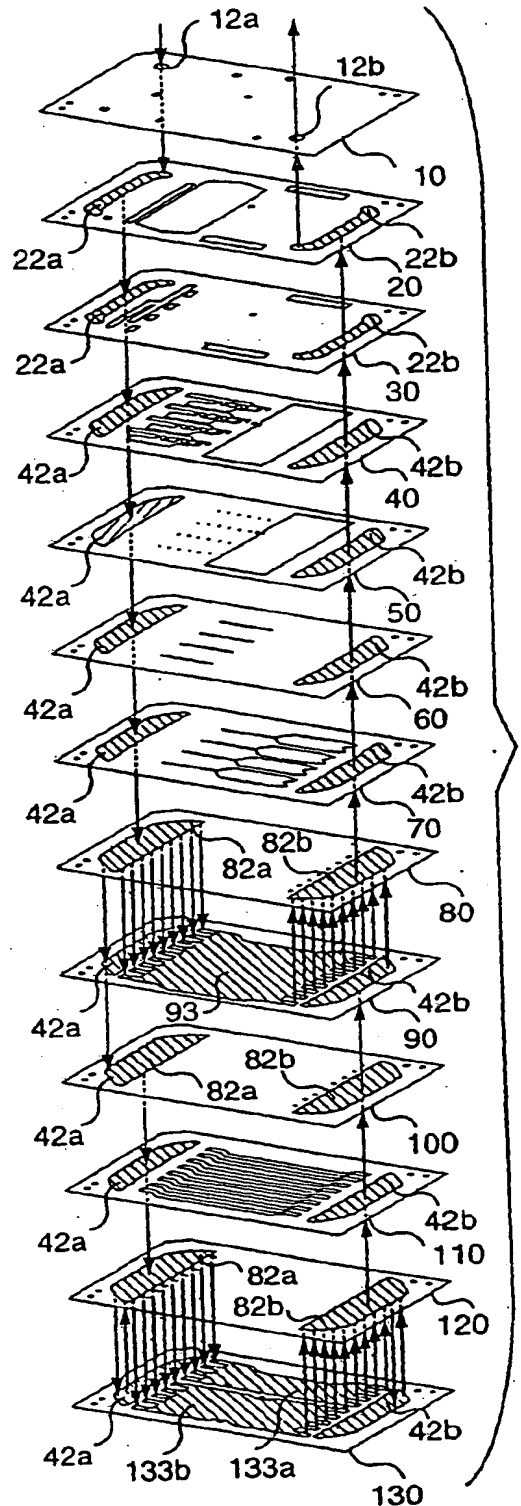


FIG. 19C



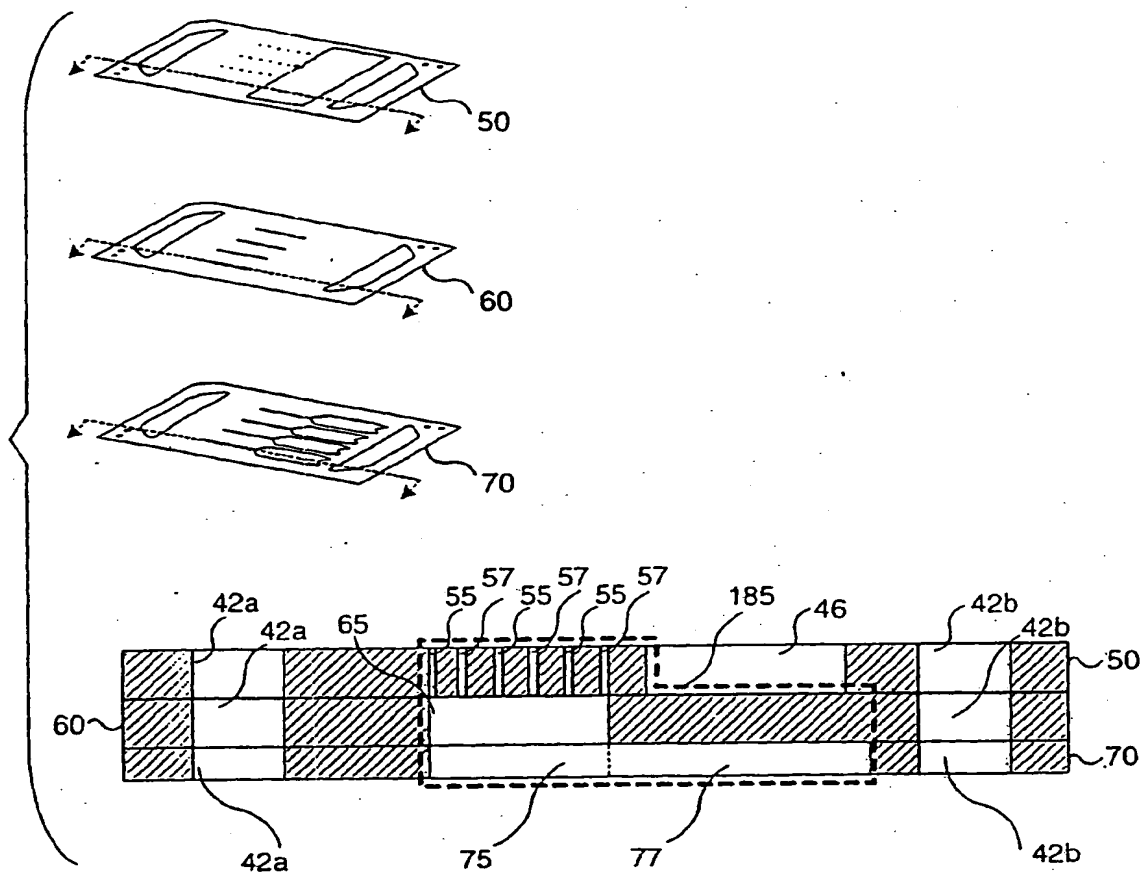


FIG. 20

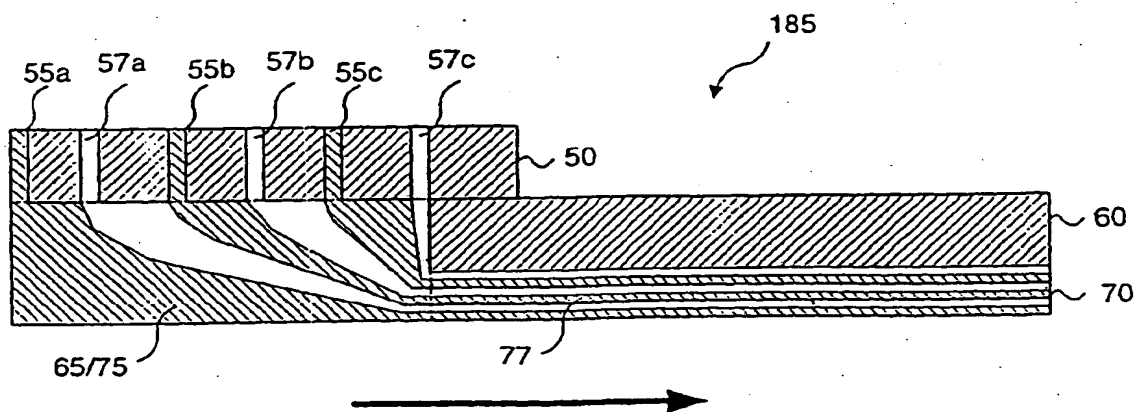
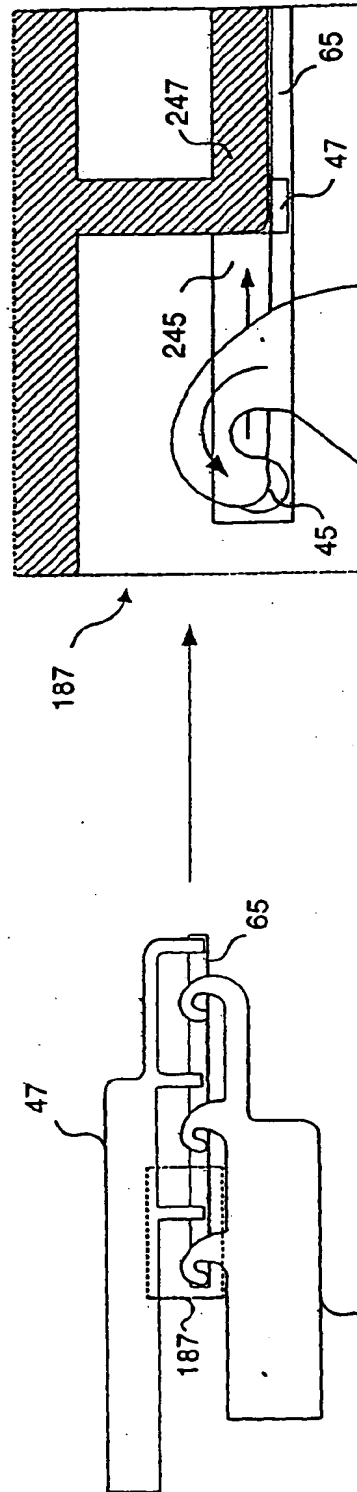
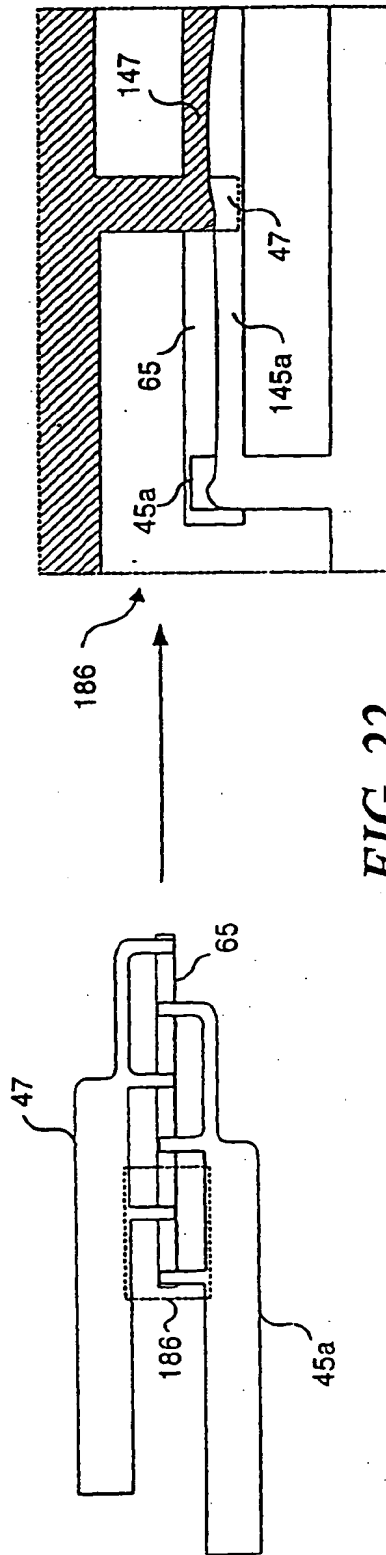
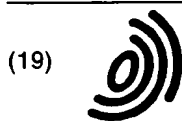


FIG. 21





Europäisches Patentamt  
European Patent Office  
Office européen des brevets



(11) **EP 1 123 734 A3**

(12)

## EUROPEAN PATENT APPLICATION

(88) Date of publication A3:  
18.12.2002 Bulletin 2002/51

(51) Int Cl.7: **B01J 19/00, B01J 19/24,  
B01F 13/00, B01F 5/06**

(43) Date of publication A2:  
16.08.2001 Bulletin 2001/33

(21) Application number: **01102270.4**

(22) Date of filing: **31.01.2001**

(84) Designated Contracting States:  
**AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU  
MC NL PT SE TR**  
Designated Extension States:  
**AL LT LV MK RO SI**

(30) Priority: **03.02.2000 US 496999**

(71) Applicant: **Cellular Process Chemistry Inc.  
Richland, Washington 99352 (US)**

(72) Inventors:  
• **Schwalbe, Thomas, Dr.  
61118 Bad Vilbel (DE)**  
• **Golbig, Klaus, Dr.  
55296 Harxheim (DE)**

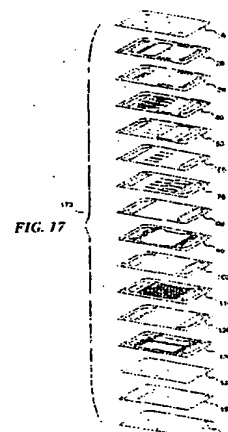
- **Hohmann, Michael, Dr.  
64287 Darmstadt (DE)**
- **Georg, Petra  
60326 Frankfurt (DE)**
- **Oberbeck, Andreas  
35781 Weilburg (DE)**
- **Dittmann, Bernd  
65843 Sulzbach (DE)**
- **Stastna, Jiri  
65760 Eschborn (DE)**
- **Oberbeck, Sebastian  
35753 Greifenstein (DE)**

(74) Representative: **Kompter, Hans-Michael  
Spessartweg 63  
64257 Darmstadt (DE)**

### (54) Miniaturized reaction apparatus

(57) A stacked plate chemical reactor in which simple plates, each incorporating no surface features other than an opening, are stacked together. When openings in adjacent plates are properly aligned, a fluid pathway is defined between inlet ports for each chemical reactant and an outlet port for a chemical product. In one embodiment of the invention, sixteen simple plates are stacked to provide a reactor incorporating three heat transfer fluid pathways, two reactant fluid pathways, one product fluid pathway, multiple mixing chambers, multiple reaction chambers, two reactant pretreatment heat exchangers, two reaction chamber heat exchangers, and multiple temperature sensor pathways. Precise dimensional control of the reactant fluid pathway height enables stacked laminar flow paths for the reactants to be achieved, allowing efficient and rapid diffusion mixing to occur. Because the simple plates incorporate no features other than openings, fabrication of such plates is easily achieved. Different reactor designs, having additional reactant pathways, more or fewer heat transfer fluid pathways, more or fewer heat exchangers, more or fewer mixing chambers, more or fewer reaction chambers, and more or fewer sensor pathways can readily be achieved by adding or removing plates from the stack, and/or by changing the pattern and number

of openings in the simple plates that are used. The simple plates can be held in the stack during use of the chemical reactor using pressure exerted on opposite outer simple plates of the stack, or can be permanently joined. A preferred material for the fabrication of the plates is stainless steel, although other materials such as glass, plastic, and other metals can alternatively be used, which are compatible with the selected reactants and the desired product.



EP 1 123 734 A3



European Patent  
Office

# EUROPEAN SEARCH REPORT

Application Number  
EP 01 10 2270

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
X	US 3 881 701 A (SCHOENMAN LEONARD, SCHINDLER ROBERT C ) 6 May 1975 (1975-05-06) * the whole document *	1-12	B01J19/00 B01J19/24 B01F13/00 B01F5/06
P, X	WO 00 34728 A (CHART MARSTON LIMITED ; SYMONDS KEITH THOMAS (GB); PELS JAN REMMERT) 15 June 2000 (2000-06-15) * page 11, paragraph 1 - page 17, paragraph 2 * * claims 1-9, 15, 16, 18-26; figures 1-8 *	1-6, 11, 12	
P, X	WO 00 62914 A (HESSEL VOLKER ; SCHIEWE JOERG (DE); EHRFELD WOLFGANG (DE); INST MIK) 26 October 2000 (2000-10-26) * page 12, paragraph 4 - page 15, paragraph 3 * * figures 1-4. *	1-3, 8-12	
A	EHRFELD W ET AL: "POTENTIALS AND REALIZATION OF MICROREACTORS", DECHEMA MONOGRAPHIEN, VERLAG CHEMIE, WEINHEIM,, DE, VOL. 132, PAGE(S) 1-28 XP000925630 ISSN: 0070-315X * page 12, paragraph 3 * * page 16, paragraph 4 - page 22, paragraph 1 *	1-12	<div>TECHNICAL FIELDS SEARCHED (Int.Cl.7)</div> <div>B01J B01F</div>
The present search report has been drawn up for all claims			
Place of search <b>THE HAGUE</b>		Date of completion of the search <b>17 October 2002</b>	Examiner: <b>Vlassis, M</b>
<b>CATEGORY OF CITED DOCUMENTS</b> X: particularly relevant if taken alone Y: particularly relevant if combined with another document of the same category A: technological background O: non-written disclosure P: intermediate document		T: theory or principle underlying the invention E: earlier patent document, but published on, or after the filing date D: document cited in the application L: document cited for other reasons &: member of the same patent family, corresponding document	

EPO FORM 503 03/82 (P04C01)



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 01 10 2270

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
T	EHRENFELD W, HESSEL V, LÖWE H: "Microreactors, New Technology for Modern Chemistry" 2000, WILEY-VCH, WEINHEIM XP002217234 * page 30, paragraph 4 - page 32, paragraph 3 * * page 35 - page 39 * * page 64, paragraph 1 - page 73, paragraph 3 * * page 83 - page 85 * * page 102, paragraph 4 - page 105, paragraph 4 * * page 112 - page 114 * -----	1-12	
			TECHNICAL FIELDS SEARCHED (Int.Cl.7)
The present search report has been drawn up for all claims			
Place of search		Date of completion of the search	Examiner
THE HAGUE		17 October 2002	Vlassis, M
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			

EPO FORM 1503 03.02 (P04C01)

**ANNEX TO THE EUROPEAN SEARCH REPORT  
ON EUROPEAN PATENT APPLICATION NO.**

EP 01 10 2270

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on  
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

17-10-2002

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
US 3881701	A	06-05-1975	NONE		
WO 0034728	A	15-06-2000	AU	1668400 A	26-06-2000
			EP	1137904 A1	04-10-2001
			WO	0034728 A1	15-06-2000
WO 0062914	A	26-10-2000	DE	19917156 A1	26-10-2000
			WO	0062914 A1	26-10-2000
			EP	1183094 A1	06-03-2002

EPO FORM P0459

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82